2007 APS USER WORKSHOP

Overcoming insufficient oversampling ratio usi ng predetermined partial information (Hard x-ray coherent diffraction imaging)

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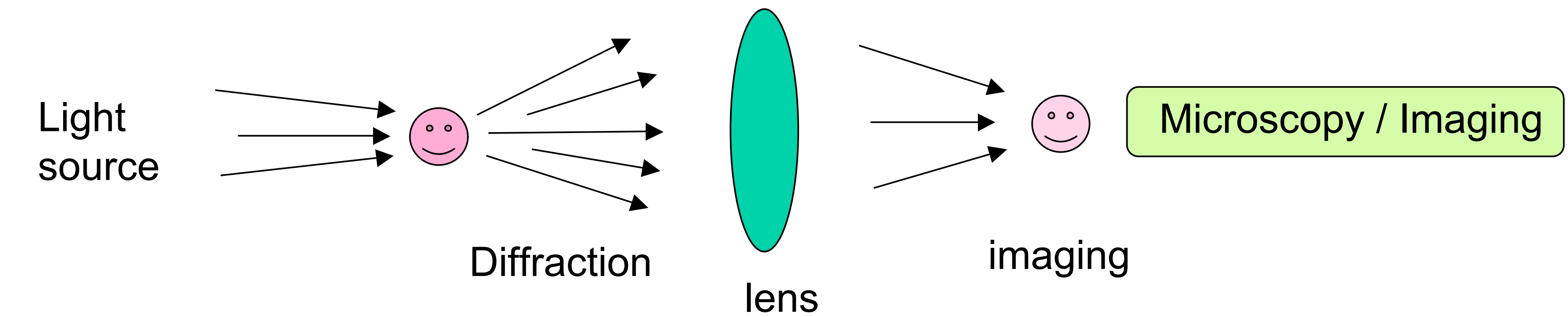
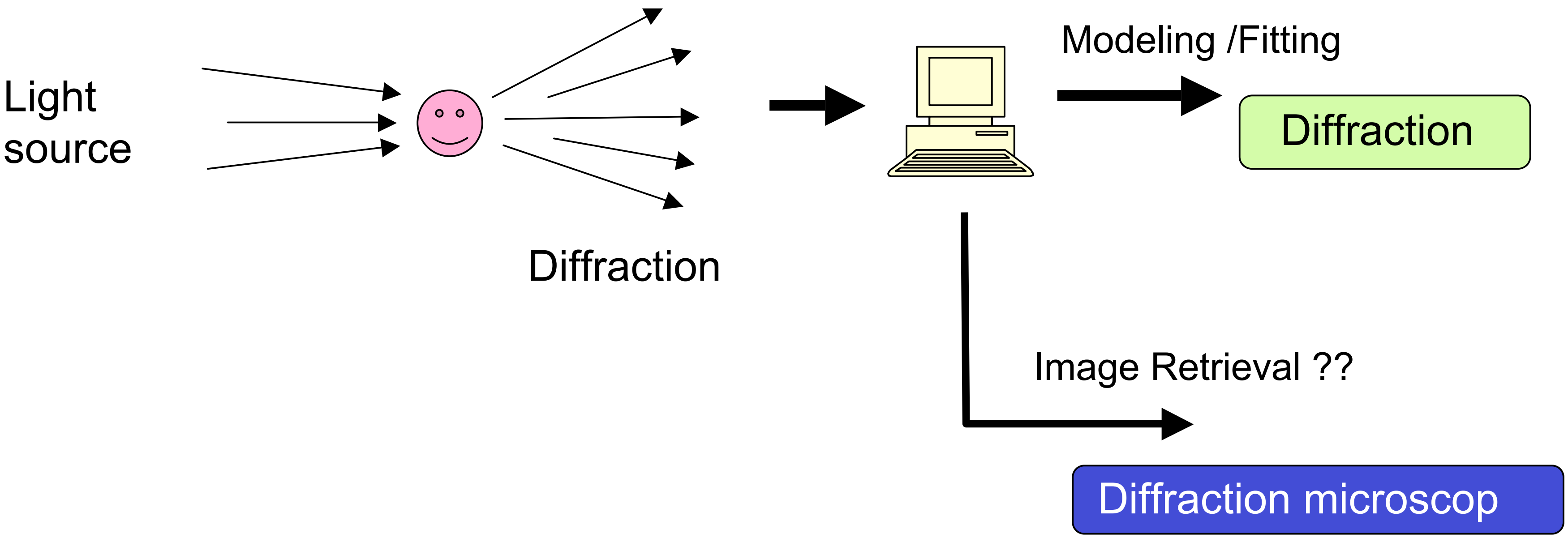
Advanced Photonic Research Institute, GIST
User Program for Ultra-Short Quantum Beam Facility

Outline

- Diffraction Imaging
- Generalized Phase Retrieval Algorithm
- Test : He-Ne diffraction
- Hard X-ray (7.5 KeV) Diffractive Imaging

Diffraction / Microscopy

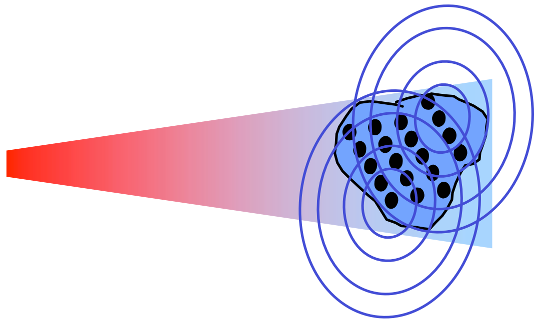




Figures from 'Element of Modern X-ray Physics'

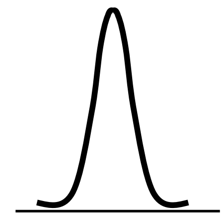
Coherent X-ray & Incoherent X-ray Diffraction

Incoherent X-ray diffraction



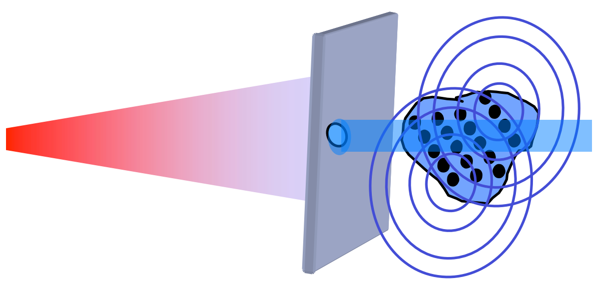
$$S(\vec{q}) = \sum_{\text{all volume}} \left| \int_{\text{coherent volume}} \rho(\vec{r}) e^{-i\vec{q}\cdot\vec{r}} \right|^2$$

$$= \left\langle \left| \int_{\text{coherent volume}} \rho(\vec{r}) e^{-i\vec{q}\cdot\vec{r}} \right|^2 \right\rangle_{\text{Average}}$$

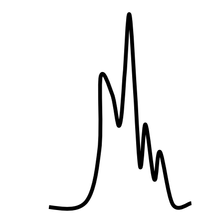


- ✓ Ensemble average of the Fourier transform inside the coherent volume
- ✓ Statistical average of the atomic scale structure

Coherent X-ray diffraction

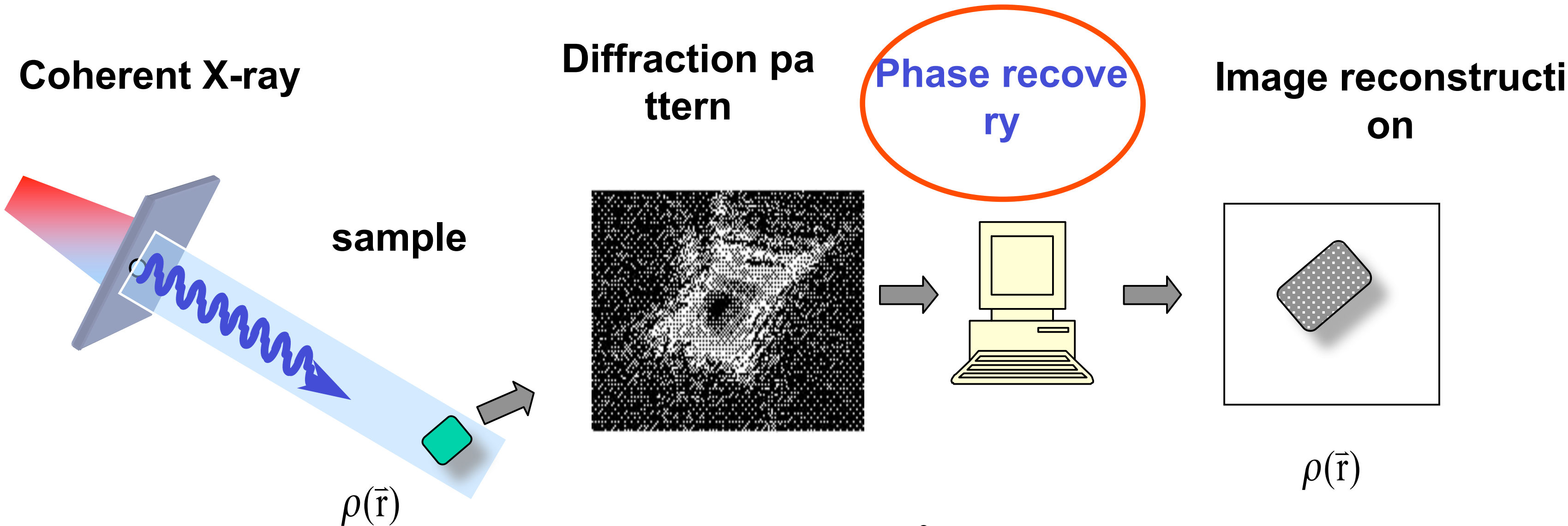


$$S(\vec{q}) = \left| \int_{\text{coherent volume}} \rho(\vec{r}) e^{-i\vec{q}\cdot\vec{r}} \right|^2$$



- ✓ Exact Fourier transform inside the coherent volume
- ✓ Individual and instantaneous structure

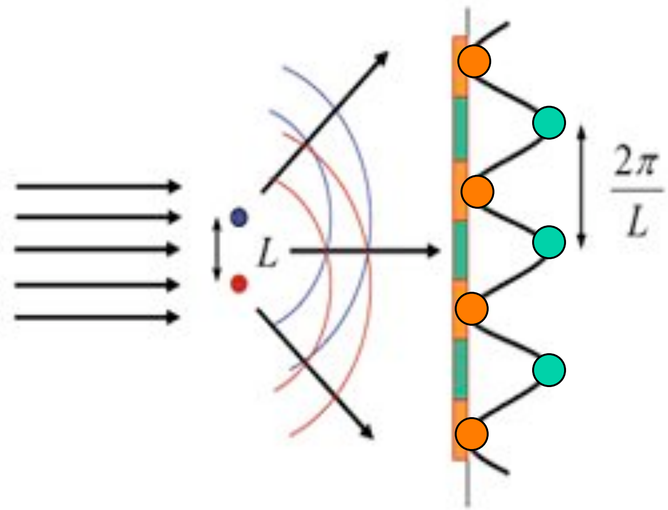
Coherent x-ray diffraction microscopy



$$S(\vec{q}) = \left| \int \rho(\vec{r}) e^{-i\vec{q}\cdot\vec{r}} \right|^2$$

- ✓ Nanometer scale resolution (ultimately atomic scale) : single particle diffraction
- ✓ Three dimensional lensless imaging
- ✓ *In-situ* non-destructive imaging

Phase Recovery : Oversampling



Sampling at frequency 2π/L is not enoug h to resolve interference fringes

$$|F(\mathbf{k})| = \left| \sum_{\mathbf{r}=0}^{N-1} \rho(\mathbf{r})\, e^{2\pi i \mathbf{k}\cdot\mathbf{r}/N} \right|$$

$$\mathbf{k} = 0,\ 1,\ 2, \ldots N-1$$

	# of unknown variables	# of independent equations
1D	2N	N
2D	$2N^2$	N^2
3D	$2N^3$	N^3

Minimum oversampling ratio is 2

J. Miao, P. Charalambous, J. Kirz, and D. Sayre, Nature 400, 342 (1999).

Oversampling Method

$$g(\mathbf{r}) = \begin{cases} \rho(\mathbf{r}) & 0 \le \mathbf{r} \le N-1 \\ 0 & N \le \mathbf{r} \le 2N-1 \end{cases}$$

Real space support constraint

$$|F(\mathbf{k})| = \left| \sum_{\mathbf{r}=0}^{2N-1} g(\mathbf{r}) e^{2\pi i \mathbf{k}\cdot\mathbf{r}/(2N)} \right|$$

Reciprocal space amplitude constraint

Oversampling ratio

$$\sigma = \frac{electron\ \ density\ \ region\ \ +\ \ no-density\ \ region}{electron\ \ density\ \ region}$$

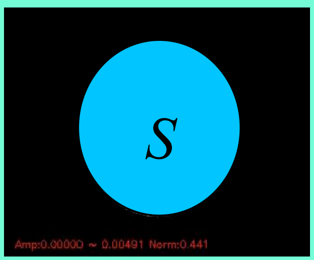
σ > 2: the phase information exists inside the diffraction intensity!

J. Miao, P. Charalambous, J. Kirz, and D. Sayre, Nature 400, 342 (1999).

Elementary Projections

Support Projection

$$\Pi_S(\vec{\rho}) = \begin{cases} \rho_r & \text{for } r \in S \\ 0 & \text{otherwise} \end{cases}$$



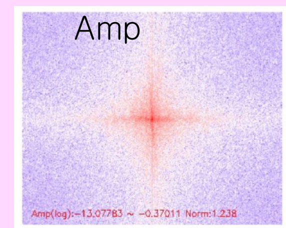
Π_S

$\Pi_S(\vec{\rho})$

$\vec{\rho}$

Fourier Modulus Projection

$$\Pi_F = F^{-1} \circ \tilde{\Pi}_F \circ F$$



phase

amplitude

$\Pi_F(\vec{\rho})$

Iterative Phase Retrieval Algorithms

$$\Pi_S[\rho(\vec{r})] = \begin{cases} \rho(\vec{r}) & for \;\; r \in S \\ 0 & otherwise \end{cases}$$

<HIO Algorithm>

$$\vec{\rho}_{n+1}(r) = \begin{cases} \Pi_F[\vec{\rho}_n(r)] & r \in S \\ \vec{\rho}_n(r) - \beta \bullet \Pi_F[\vec{\rho}_n(r)] & r \notin S \end{cases}$$

J.R. Fienup, Appl. Opt. 21, 2758 (1982)

<Difference Map>

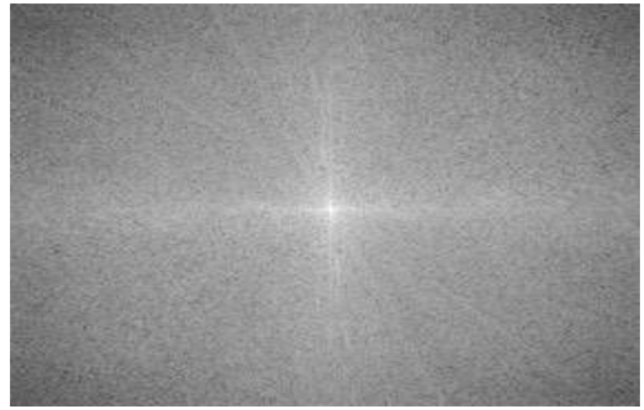
$$\vec{\rho}_{n+1} = \vec{\rho}_n + \beta \Delta \vec{\rho}_n$$

$$\Delta = \Pi_S \circ [(1+\gamma_2)\Pi_F - \gamma_2] - \Pi_F \circ [(1+\gamma_1)\Pi_S - \gamma_1]$$

J.R. Fienup and C.C. Wackerman, J. Opt.Soc.Am. A3,1897(198
6) .
J.R. Fienup, J. Opt. Soc.Am. A4,118(1987).

Test of Phase Retrieval Algorithm

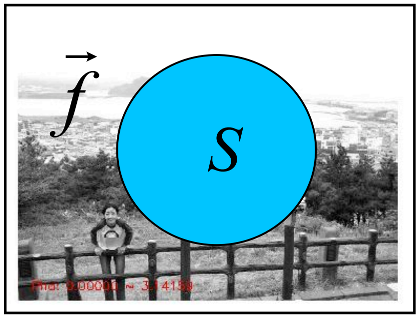
2D



Generalized Phase Retrieval Algorit hm

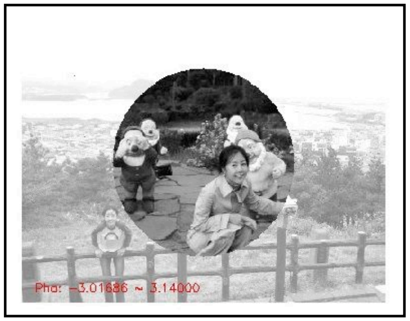
Generalized Support Projection

$$\Pi_{S(\vec{f})}(\vec{\rho}) = \begin{cases} \rho_r & \text{for} \quad r \in S \\ f_r & \text{for} \quad r \notin S \end{cases}$$



$$\Pi_{S(\vec{f})}$$

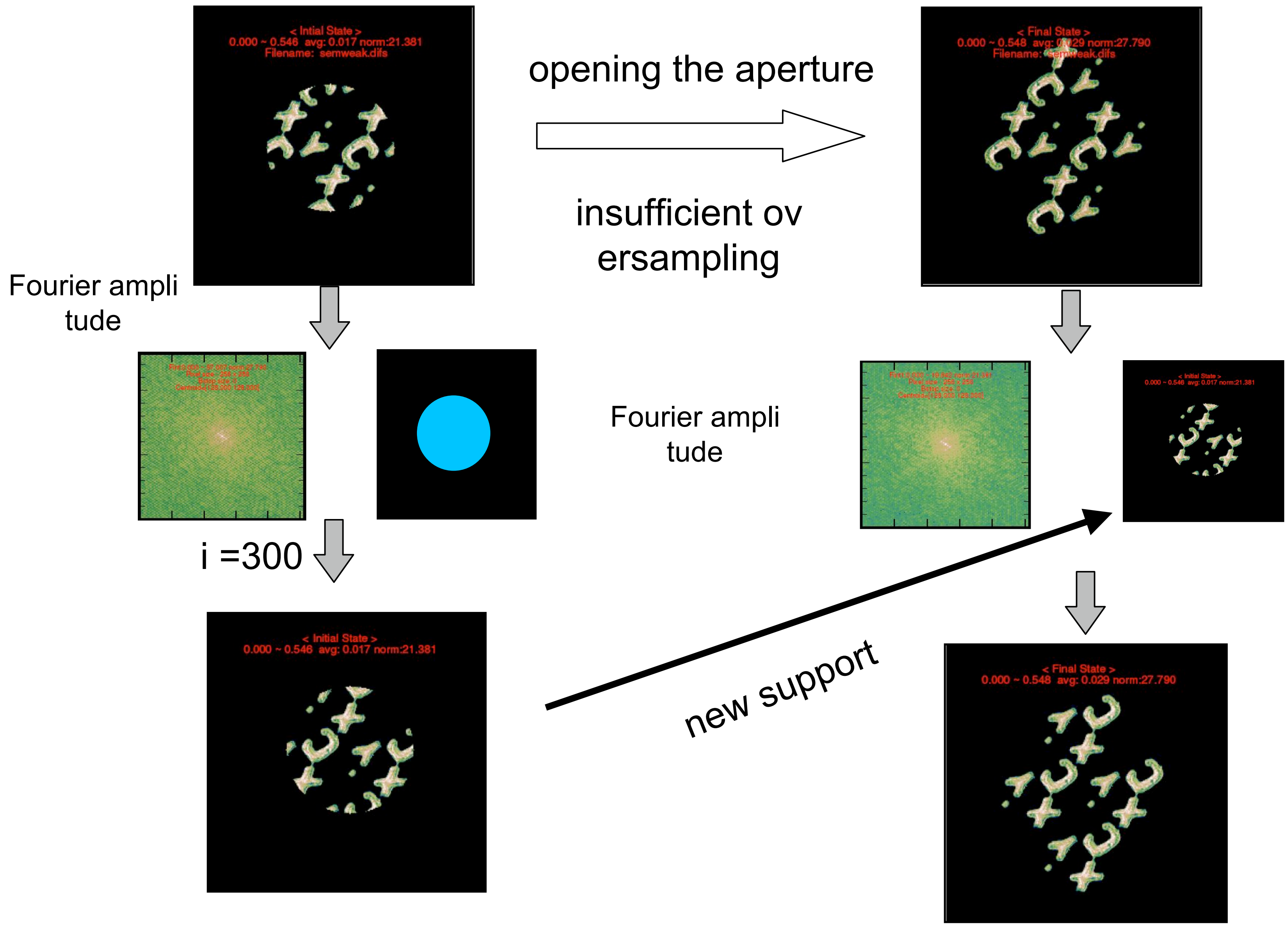
projection



$\vec{\rho}$

$\Pi_{S(\vec{f})}(\vec{\rho})$

Phase Retrieval using predetermined information



Generalized Iterative Phase Retrieval Algorithms

$$\Pi_S^{\vec{K}}[\rho(\vec{r})] = \begin{cases} \rho(\vec{r}) & for \;\; r \in S \\ K(\vec{r}) & otherwise \end{cases}$$

<HIO Algorithm>

$$\vec{\rho}_{n+1}(r) = \begin{cases} \Pi_F[\vec{\rho}_n(r)] & r \in S \\ \vec{\rho}_n(r) + \beta\{\vec{K}(r) - \Pi_F[\vec{\rho}_n(r)]\} & r \notin S \end{cases}$$

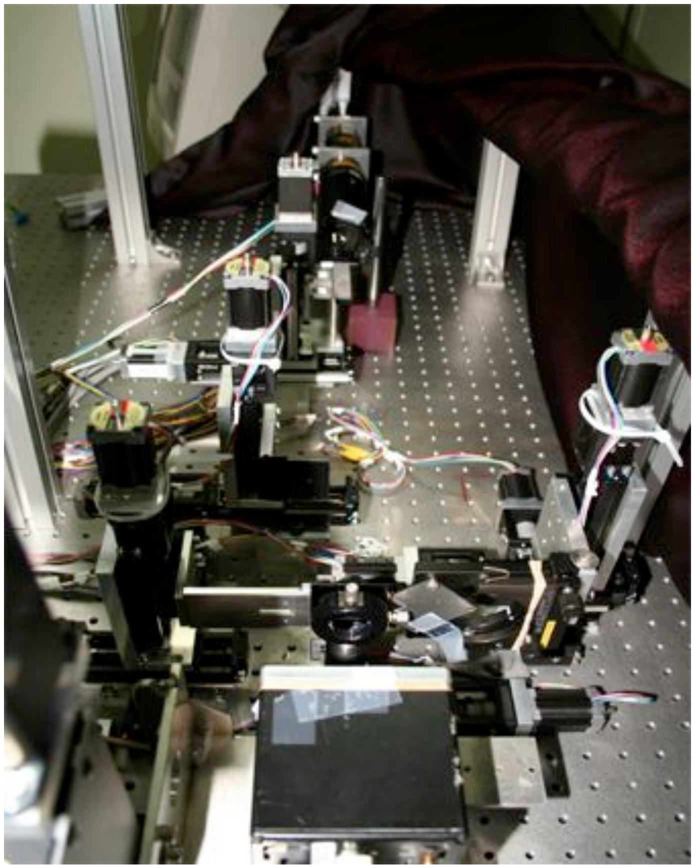
<Difference Map>

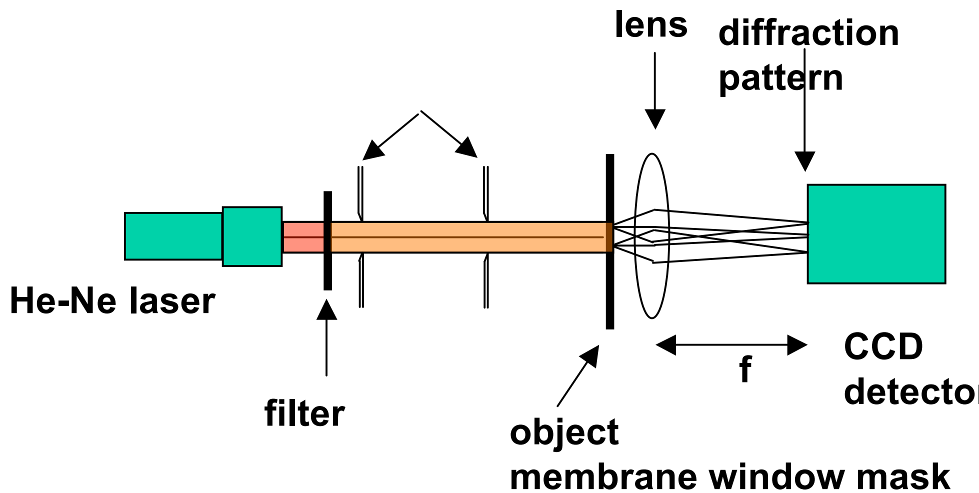
$$\vec{\rho}_{n+1} = \vec{\rho}_n + \beta\Delta\vec{\rho}_n$$

$$\Delta = \Pi_S^{\vec{K}} \circ [(1+\gamma_2)\Pi_F - \gamma_2] - \Pi_F \circ [(1+\gamma_1)\Pi_S^{\vec{K}} - \gamma_1]$$

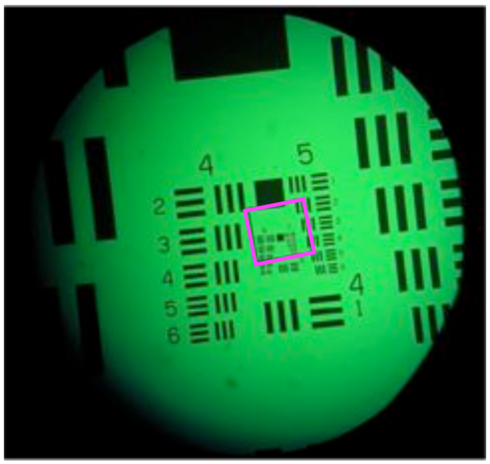
Test of the Generalized Phase Retri eval Algorithm with He-Ne Laser Diffraction

He-Ne Laser Experimental Setup

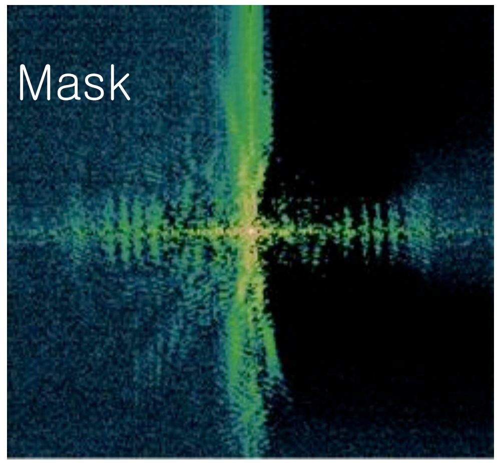




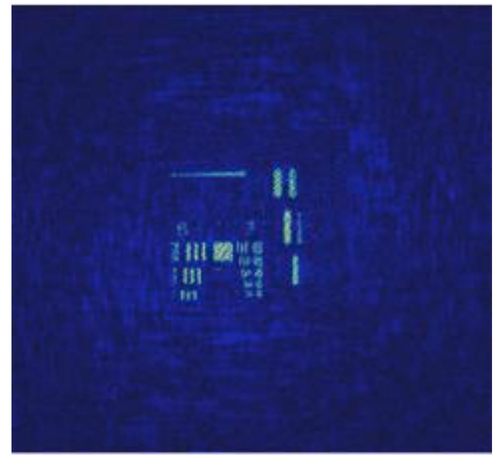
Typical Diffraction & Imaging



Original Image

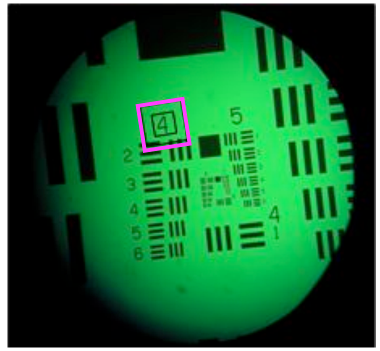


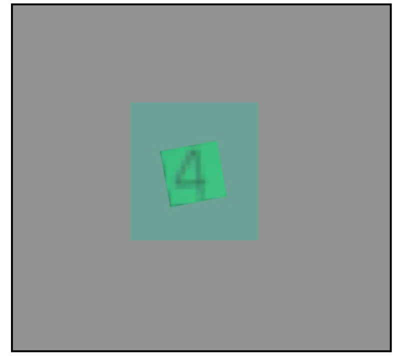
Diffraction Pattern

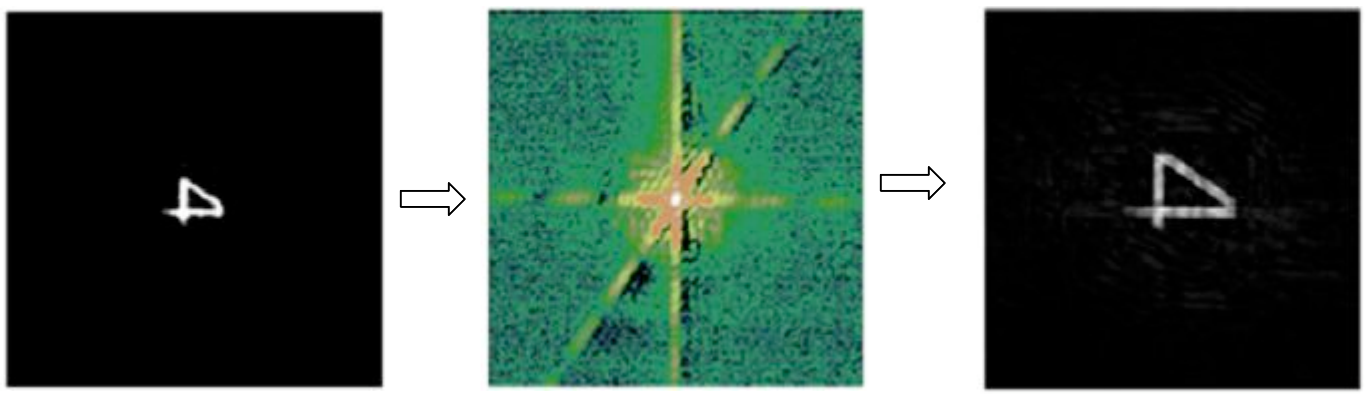


Reconstructed Image

Reconstruction







Optical Image Diffraction Pattern Image reconstruction

Reconstruction Using Predetermined Information

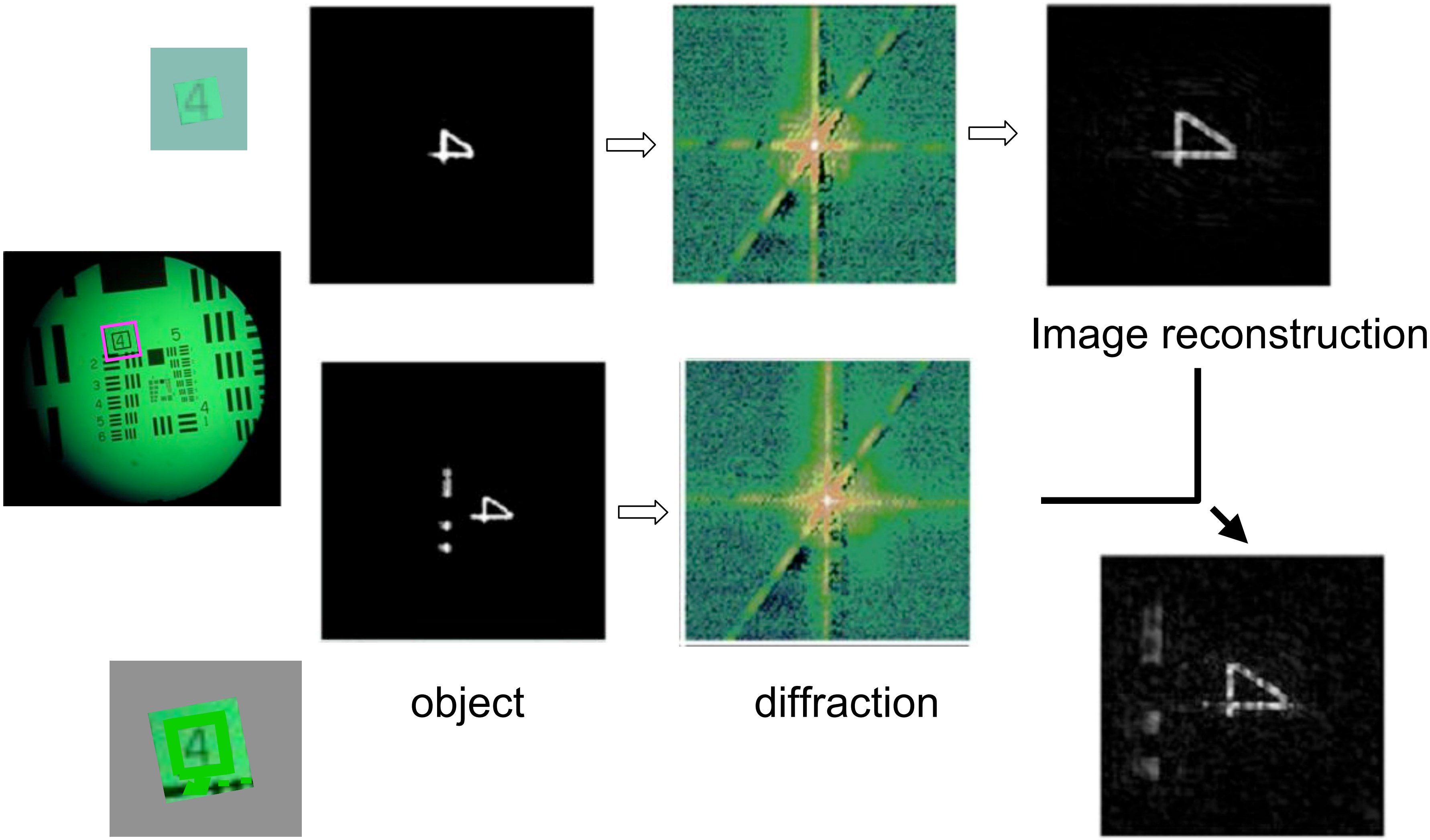
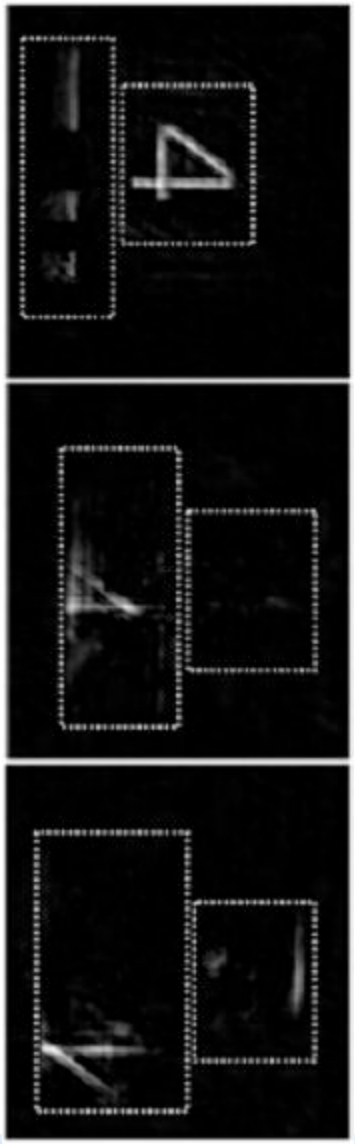


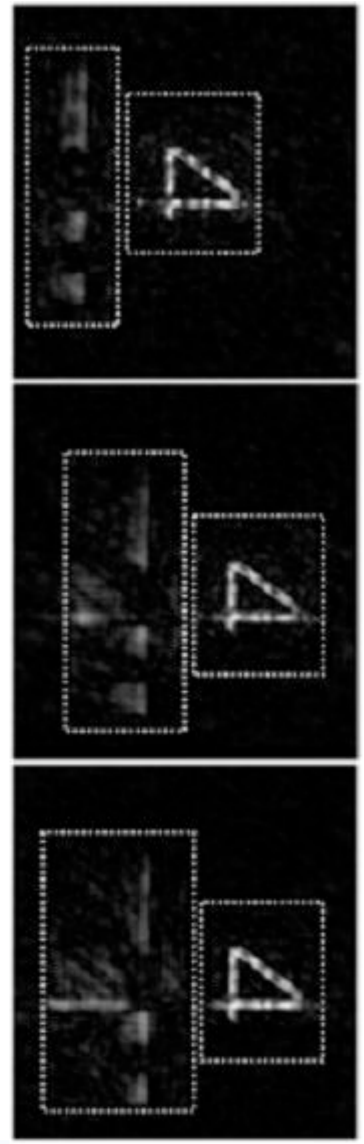
Image reconstruction with predetermined information

Dependence on Oversampling Ratio

Ordinary oversampling

Generalized oversampling





σ = 3.0

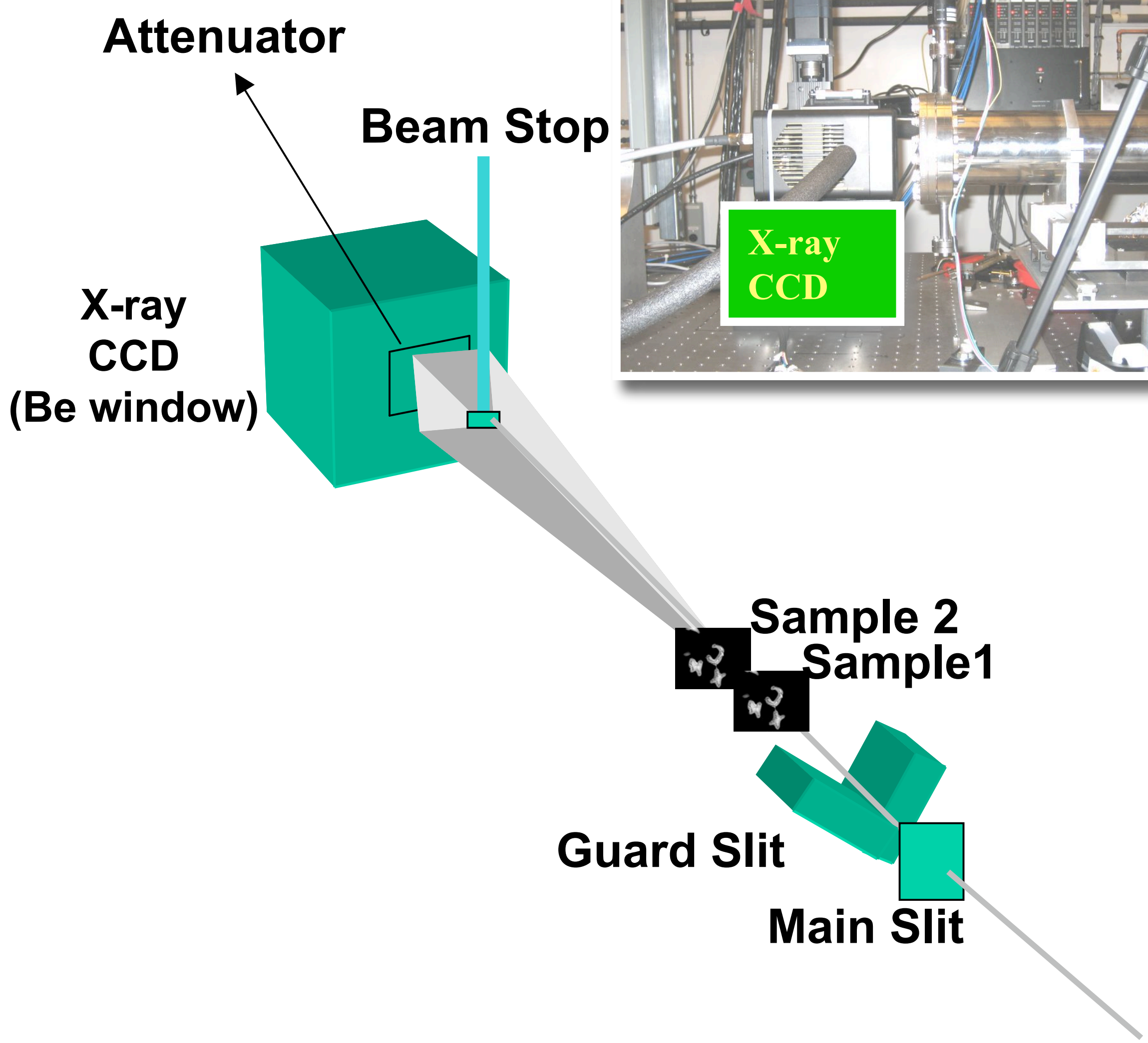
σ = 2.5

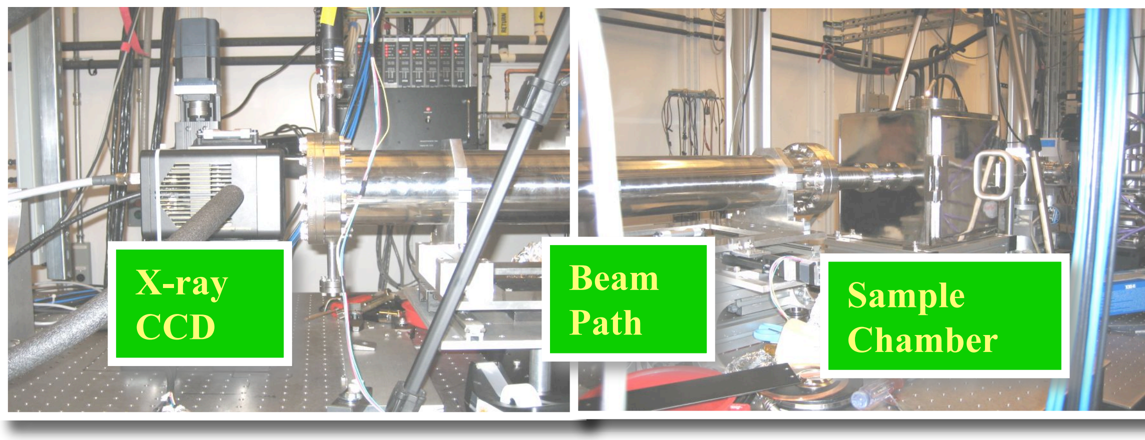
σ = 2.0

Hard x-ray coherent diffraction imag ing

Advanced Photon Source

Schematic Setup (APS-8ID)

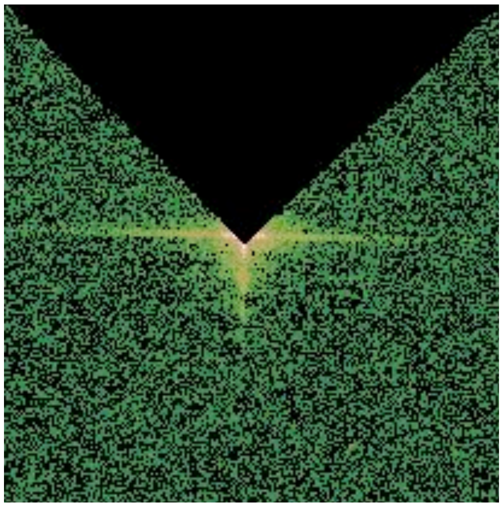




E=7.45 keV

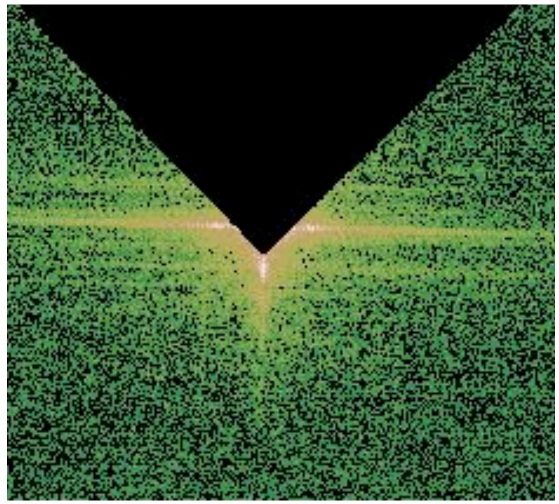
λ=0.17 nm

Central Pixels : Attenuators



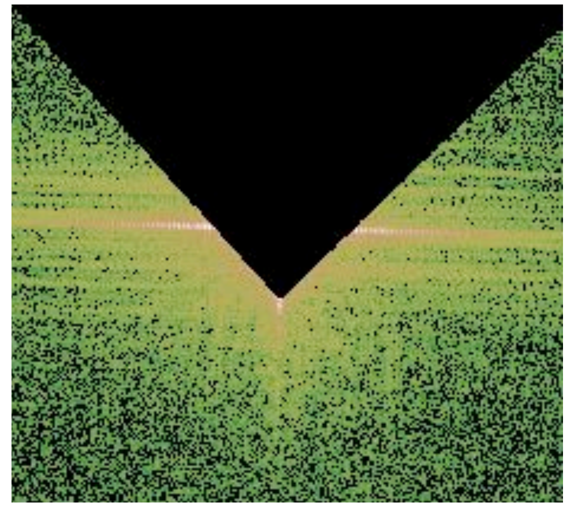
Al :360 μm

Atten. factor: 0.0037



Al :160 μm

Atten. factor: 0.083



no attenuator

CCD Model: PI-LCX1300(Be window type)

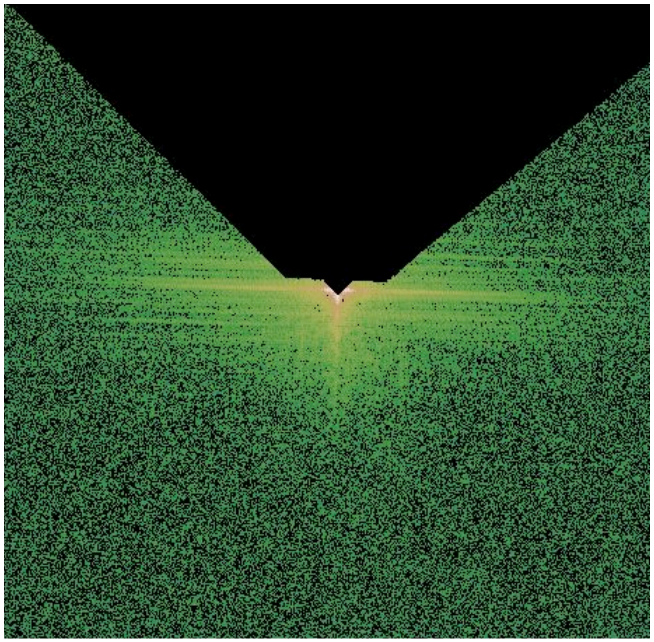
Attenuator : Al foils for UHV purpose.

Total Expose time: 100s

(200 frames with expose time,0.5s, were gathered safely)

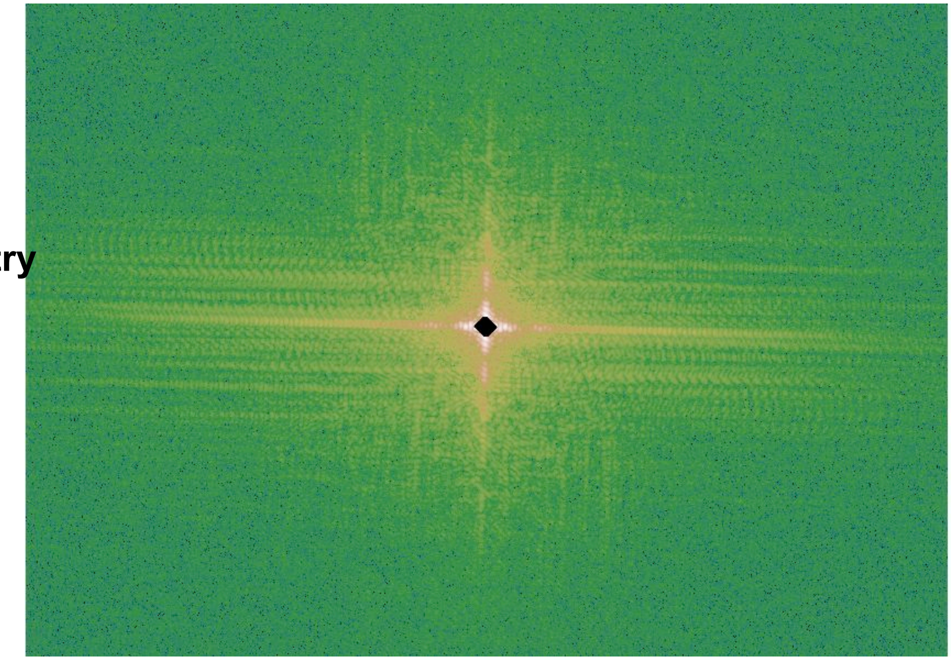
DATA PROCESSING

Merged Data



Centrosymmetry

Final Data

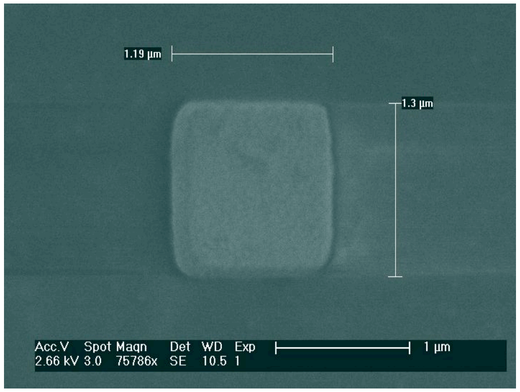


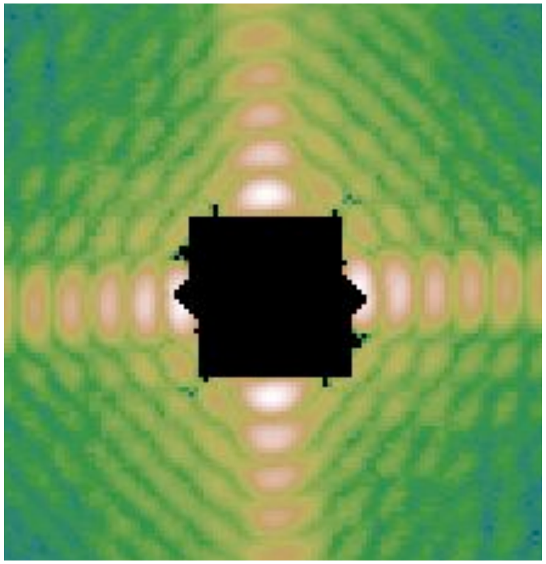
Choose the maximum intensity direction and divide the corresponding factor

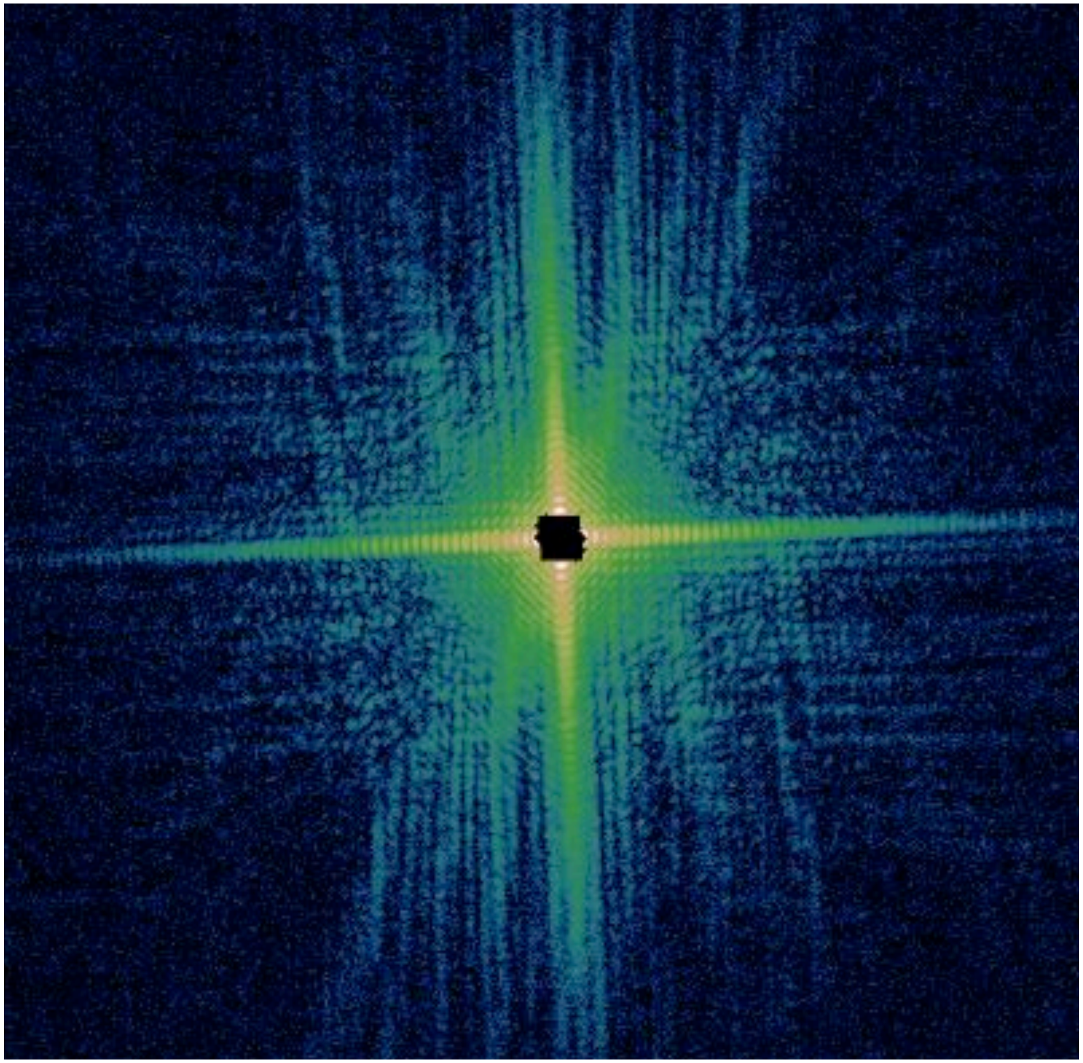
Sampling distance in real space ~ 9.3 ㎛

Q range: ~ 0.061 Å^{-1}

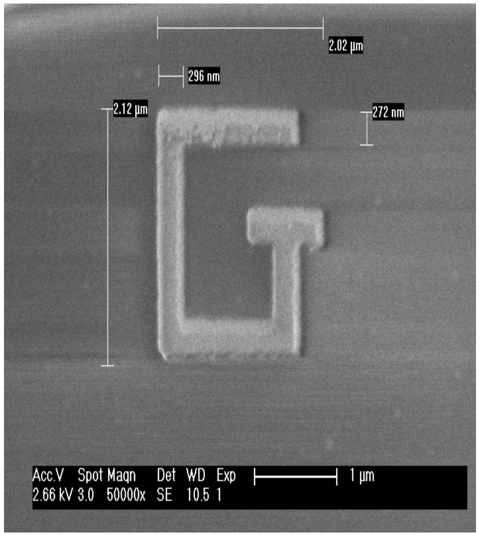
Test Pattern 1: Gold Square

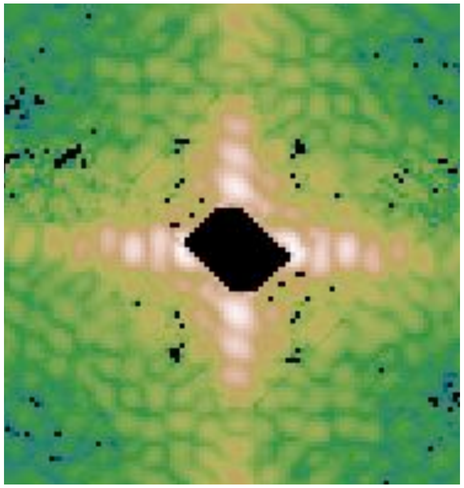






Gold Letter : G





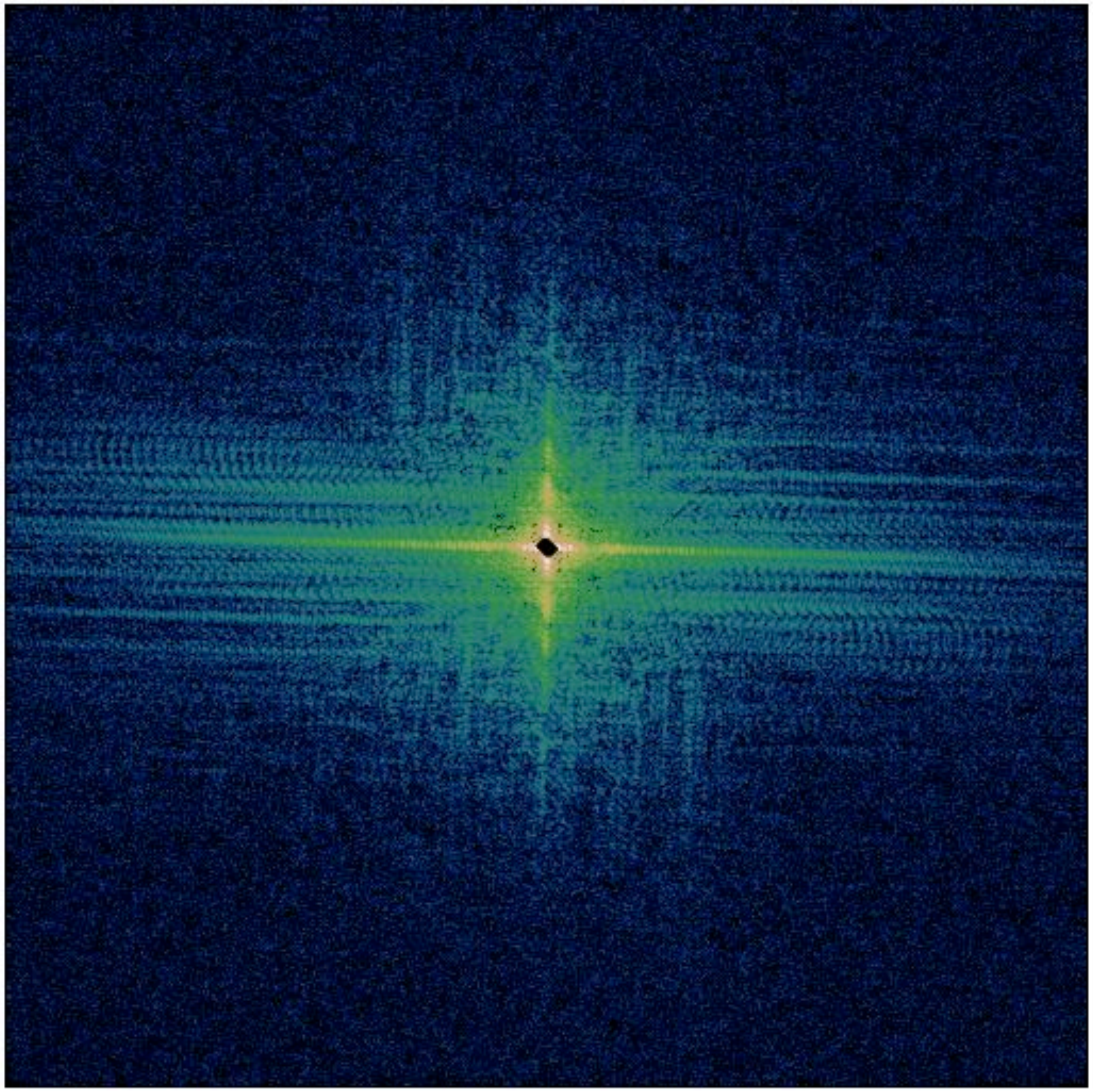
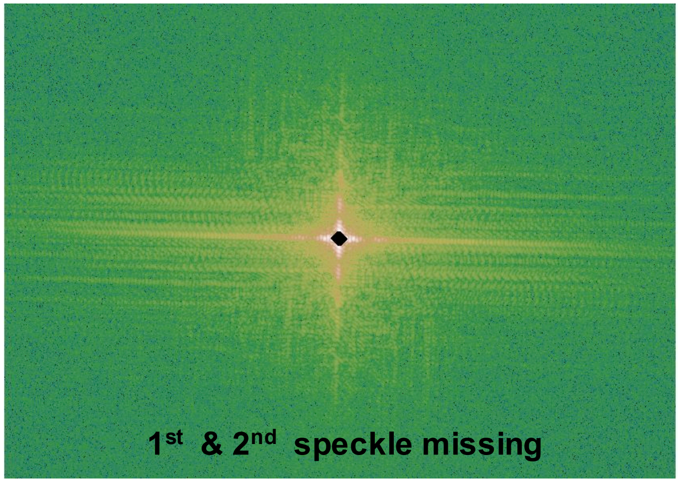
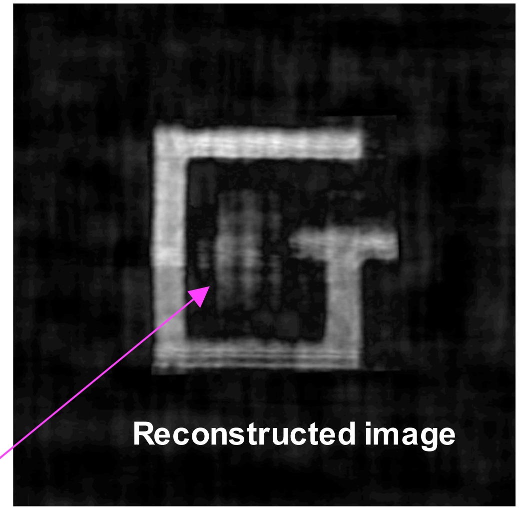


Image Reconstruction(HIO)





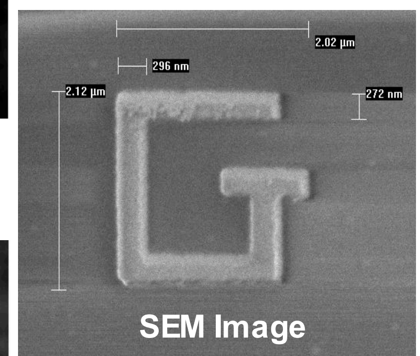
HIO Method

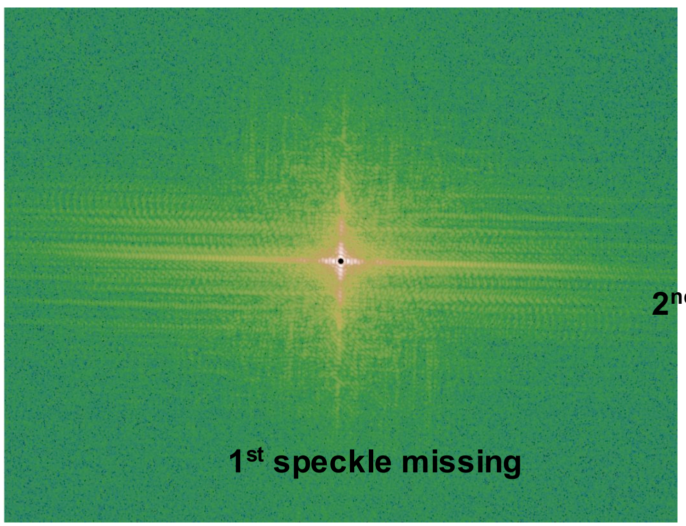
Iteration~ 3000

Err ~ 0.1

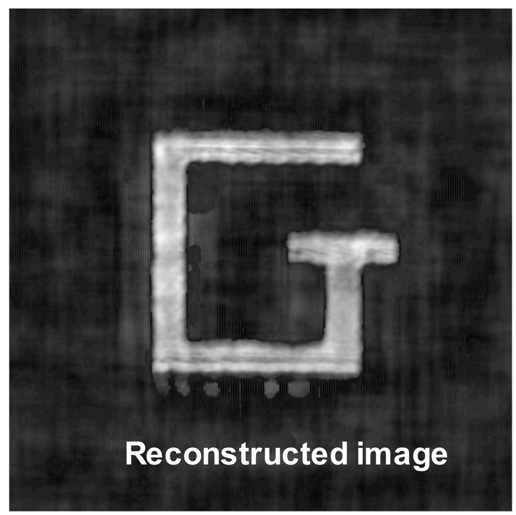
Artifact comes from

the missing 2nd speckle





2nd speckle patc hed



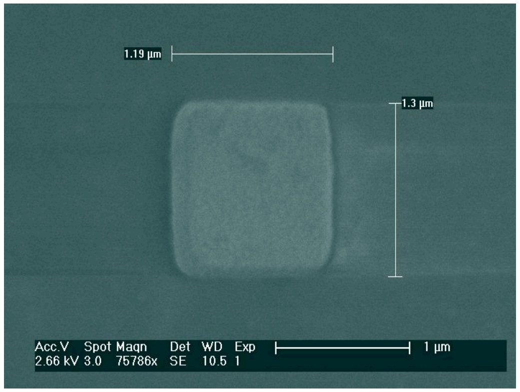
HIO Method

Iteration~ 3000

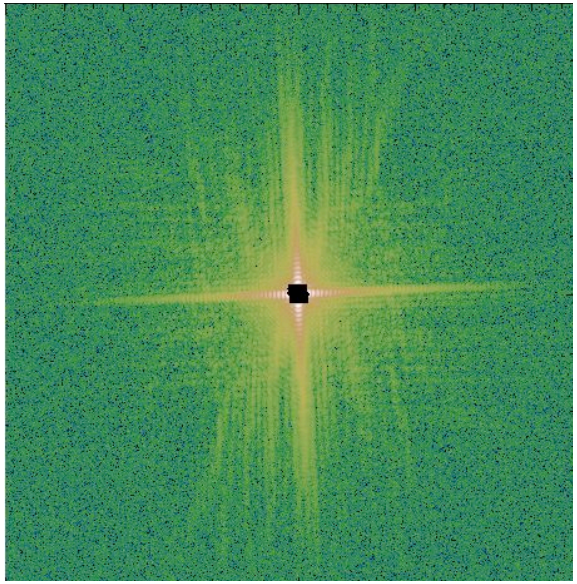
Err ~ 0.05

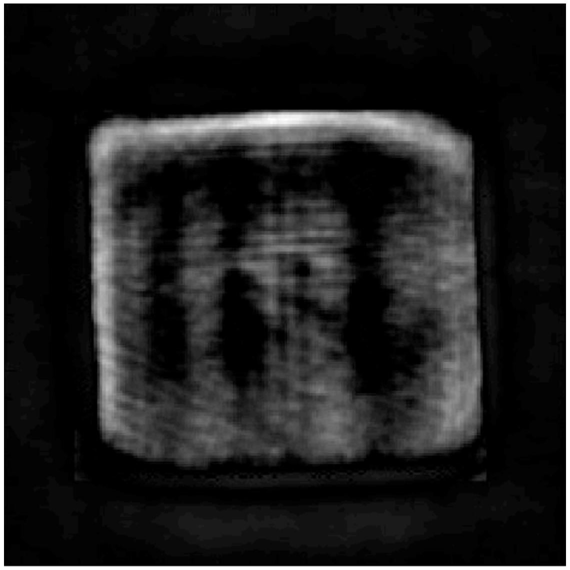
Missing data -> using Y.Nishino method (Y.Nishino et al. Phys.Rev.B 68,220101(2003)

Image Reconstruction



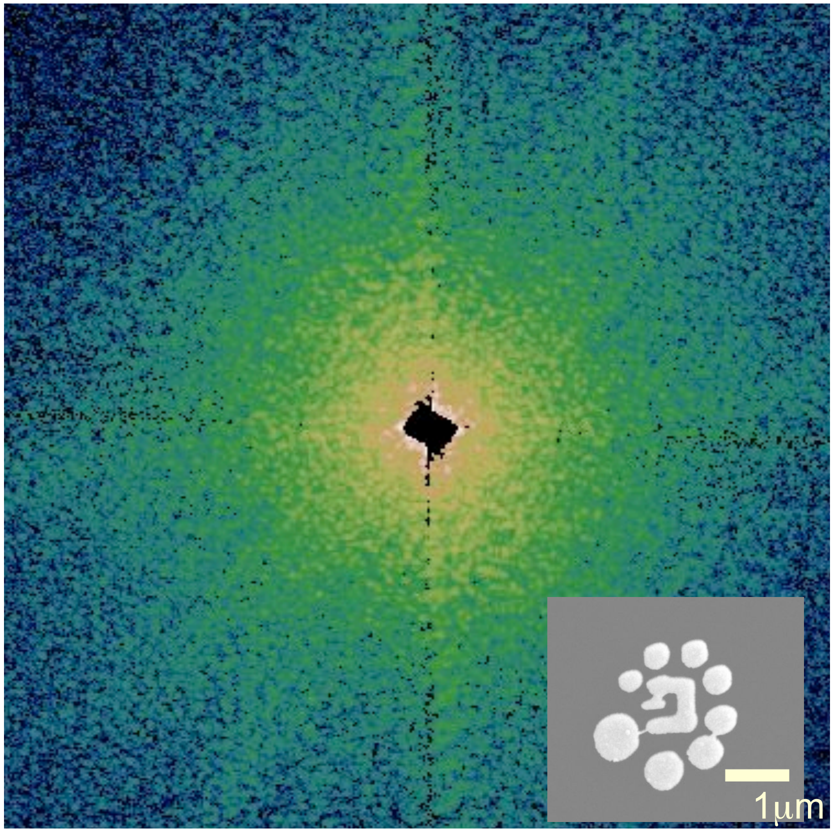
(SEM)

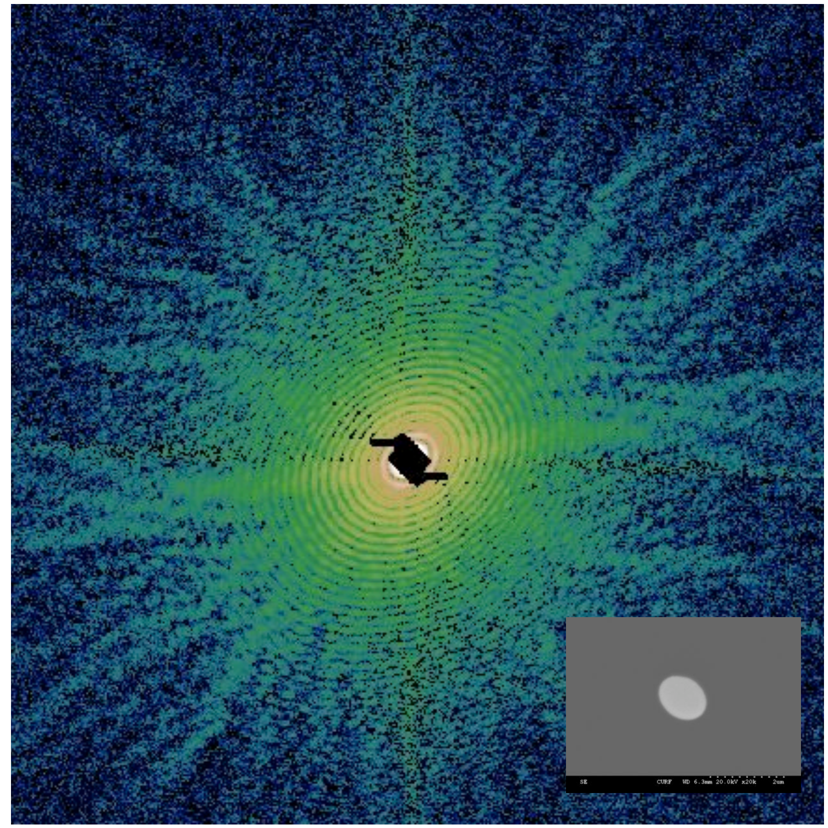




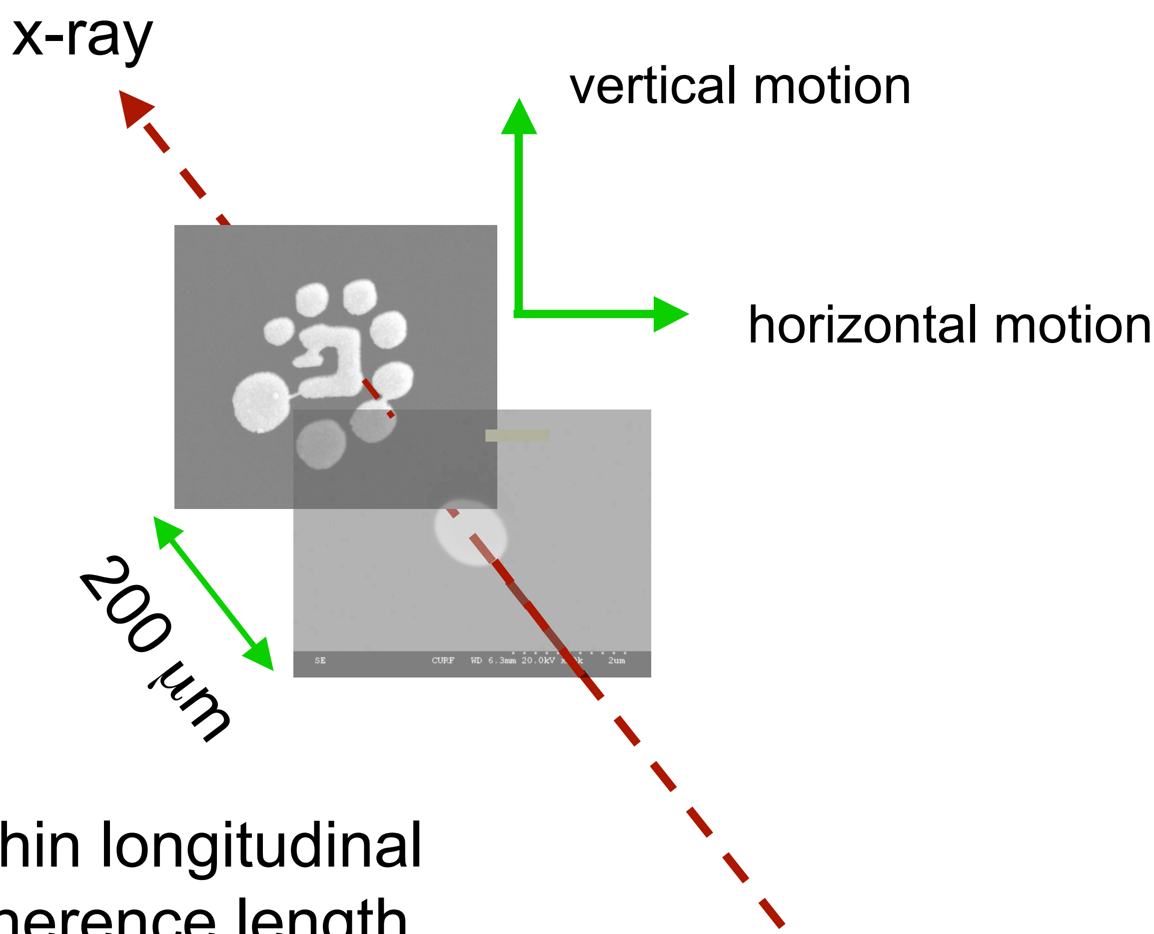
(Reconstructed)

Testing the generalized algorithm with hard x-rays





Sample Geometry



within longitudinal
coherence length

Diffraction pattern of an ellipse

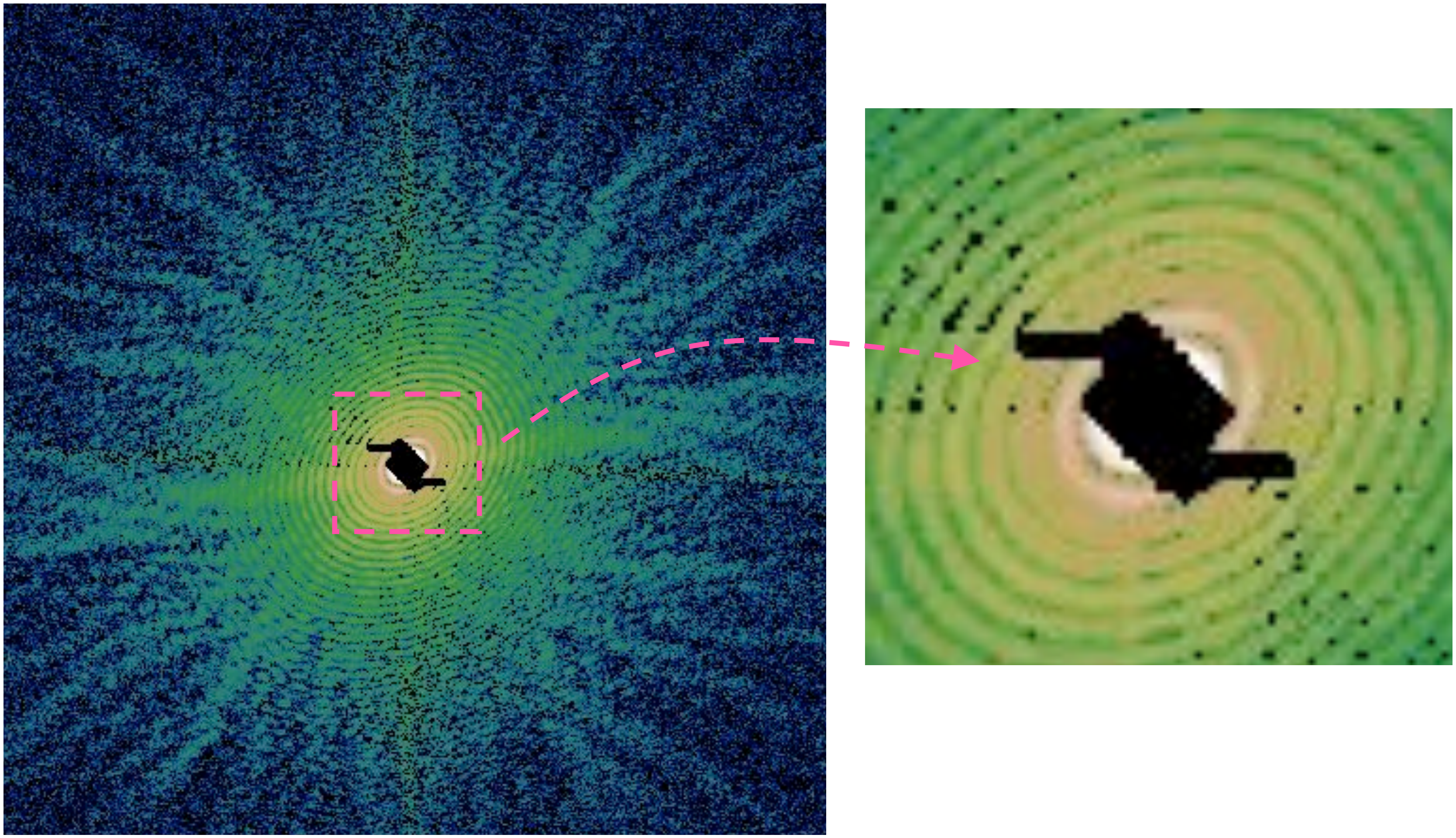
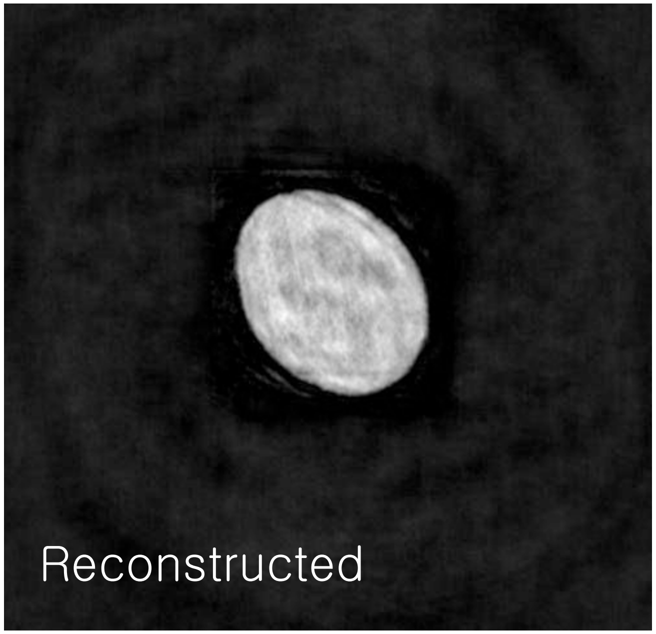
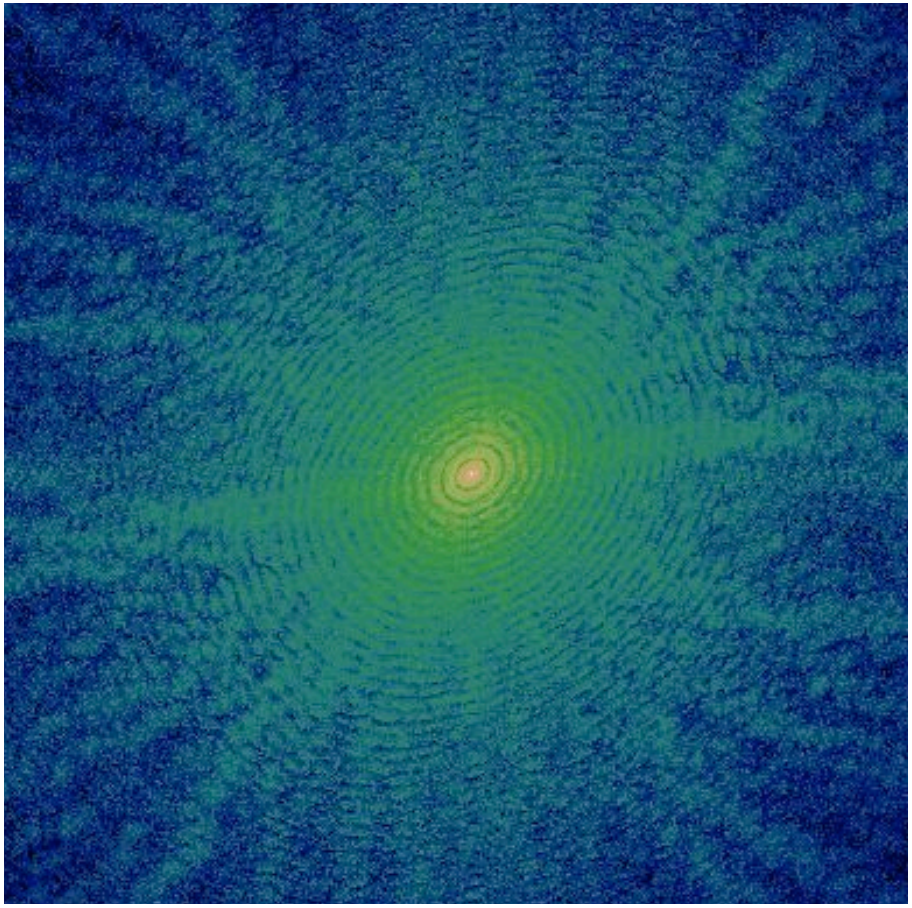


Image Reconstruction of an ellipse



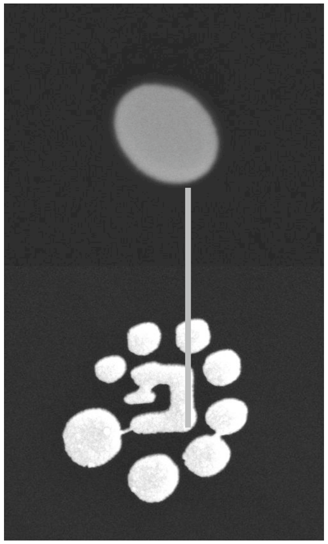
Reconstructed Image

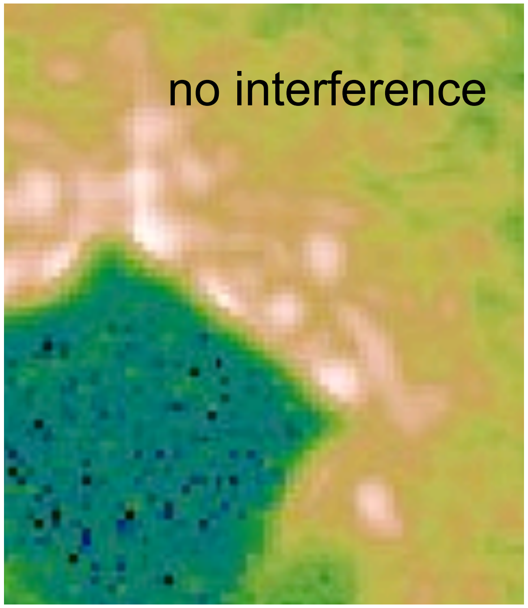
(HIO)

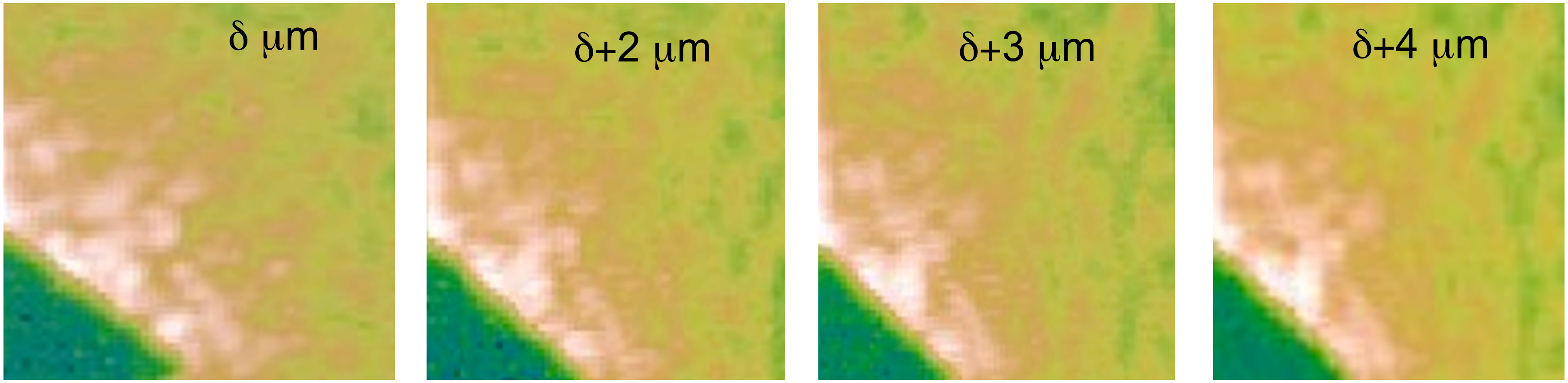


Calculated diffraction pattern

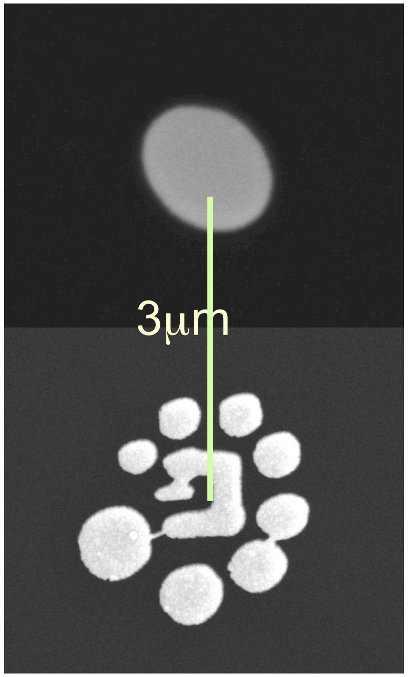
Two particle interference pattern : Hard x-ray

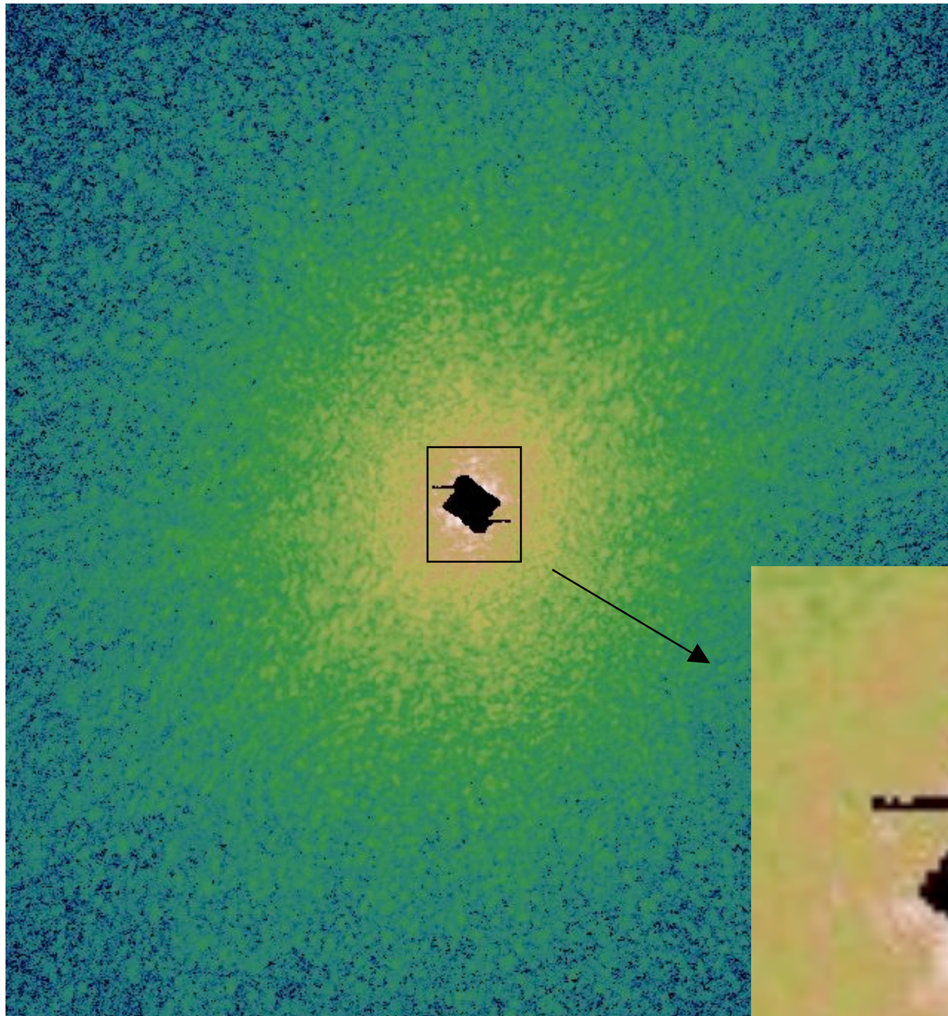




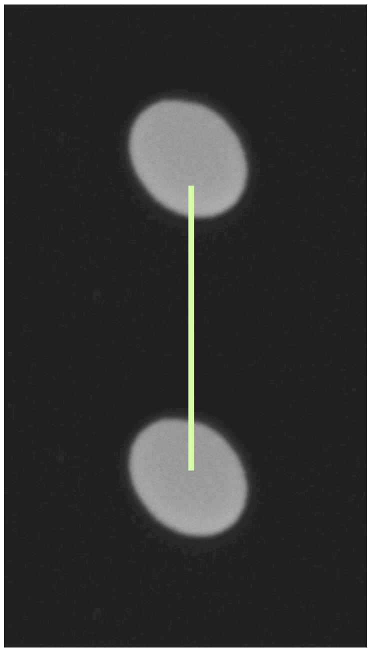


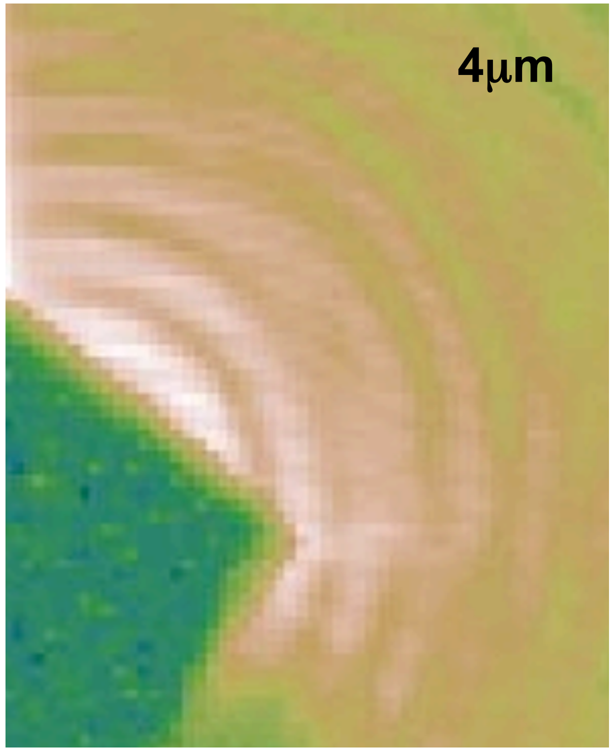
Testing the generalized algorithm (on going)

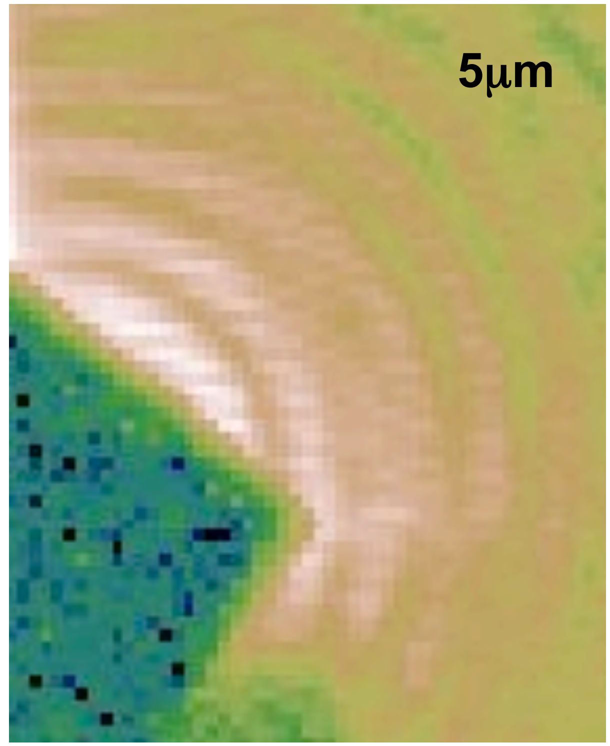




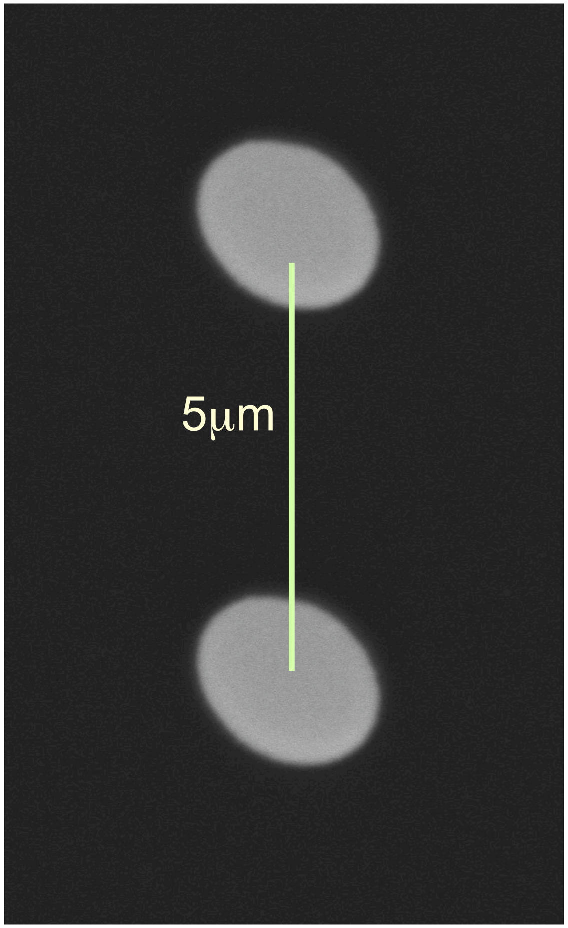
Two ellipse interference pattern : Hard x-ray

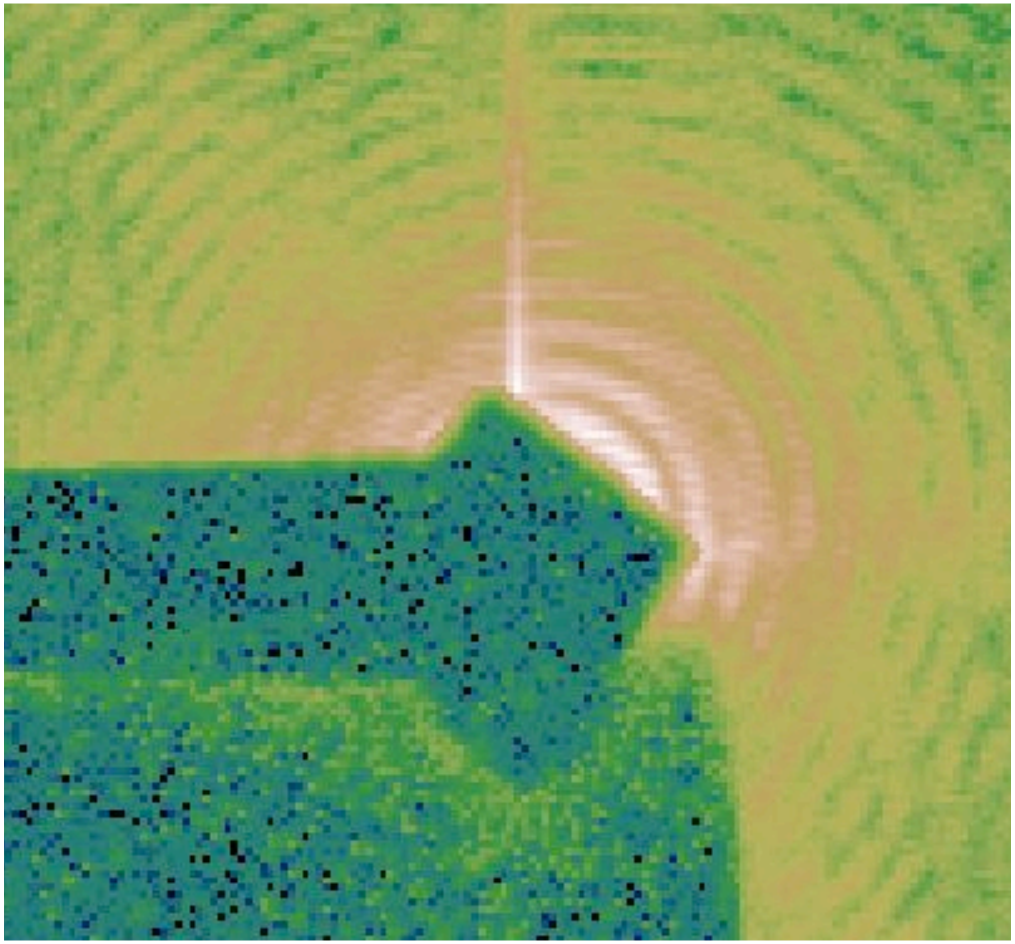




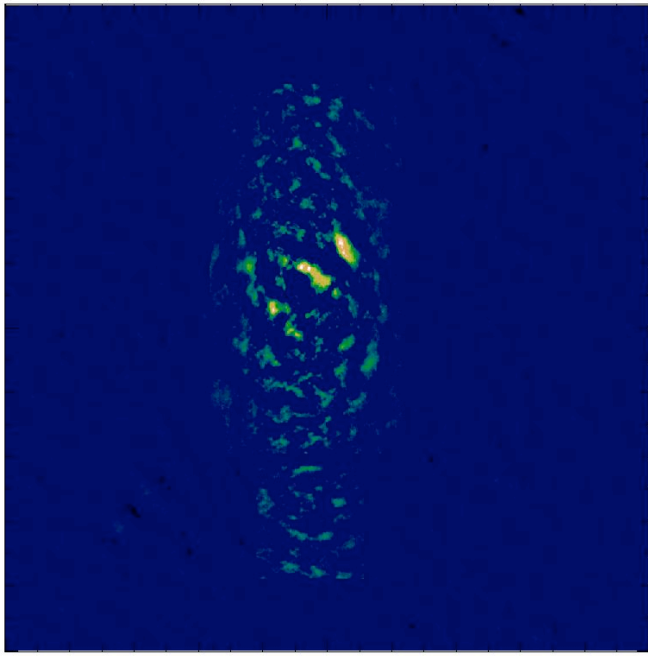


Testing the generalized algorithm

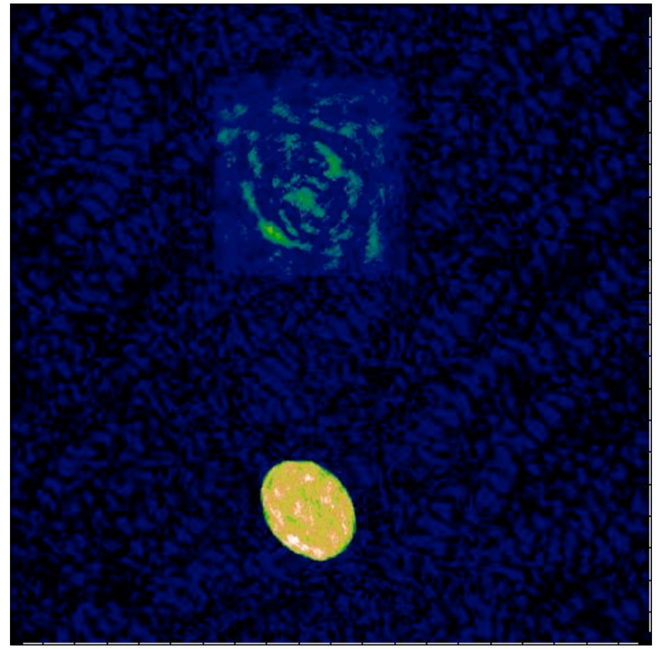




Ordinary versus generalized algorithm

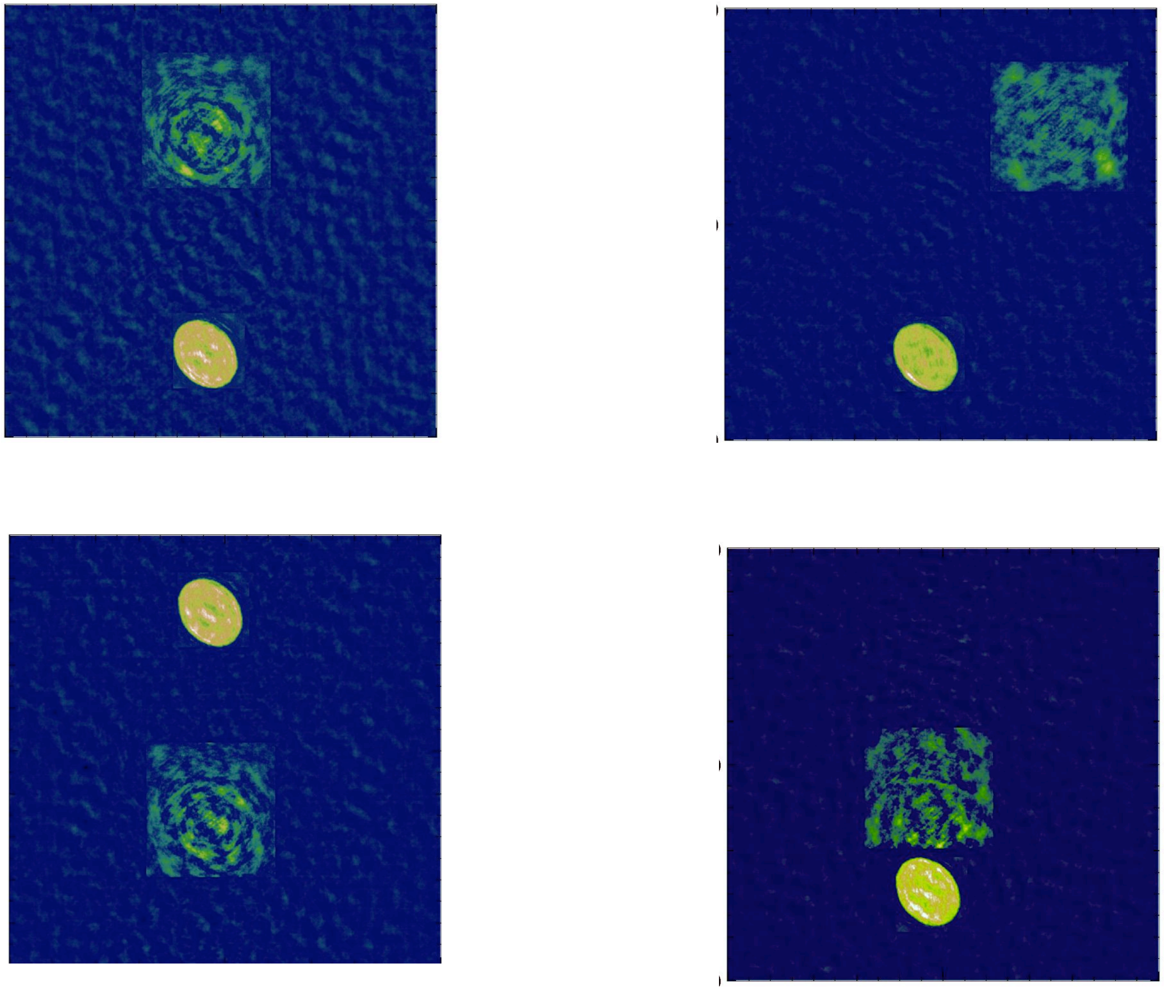


Direct HIO



Generalized HIO

Testing the generalized algorithm



Prospects of hard x-ray coherent diffraction imaging

Bright sides

- High resolution : wavelength of order 0.15 nm
  (single molecular structure often require resolution below 1 nm)
- Coherency : transverse coherence of order 10 μm
  (in most cases, field of view is less than 5 μm)
- Non vacuum applications (in-situ, in-vivo, …)

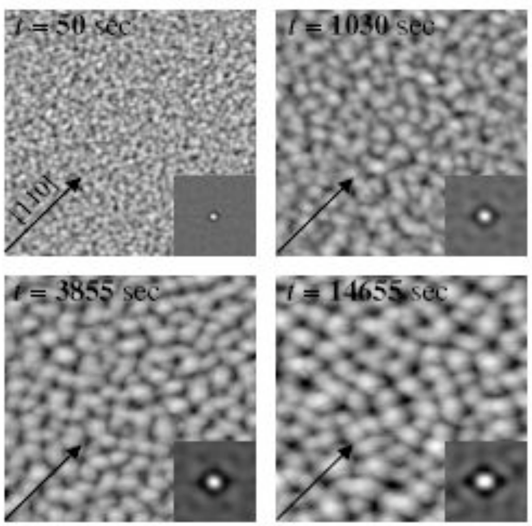
Limiting factors

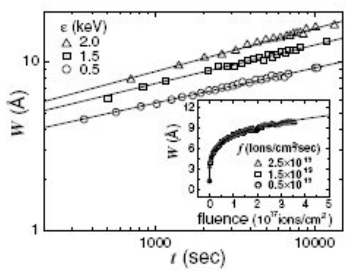
- Need higher flux x-ray sources → Future x-ray sources
- CCD detectors of large dynamic ranges are required
- Sample handling & environments (Dedicated beamlines …)

Applications of hard x-ray coherent diffraction imaging

Applications in nanotechnology (inorganic/organic)

Reflection geometry is required
Non-isolated specimen (x-ray pinhole)
Buried Interface structure (may require sample thinning)





surface science

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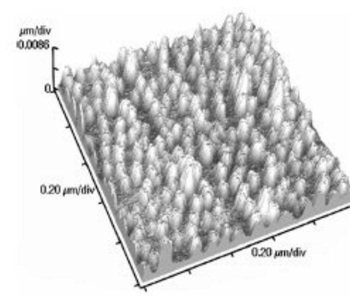
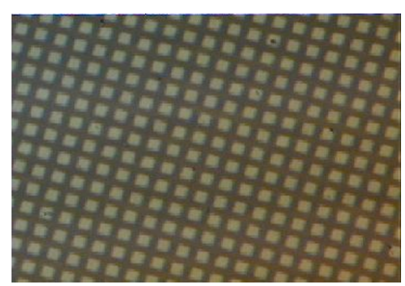
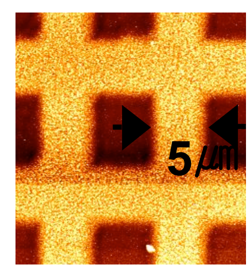


Fig. 1. AFM image of uncapped $In_{0.5}Ga_{0.5}As$ quantum dots formed on GaAs(0 0 1) surface.

Quantum dots



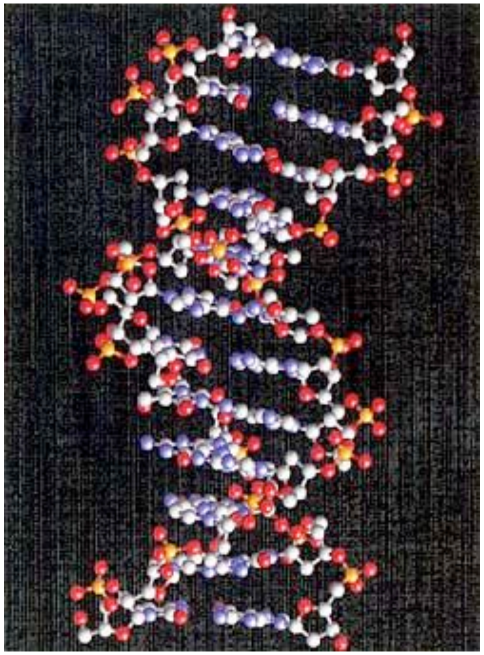


nano structures

Applications of hard x-ray coherent diffraction imaging

- Molecular level technology (single molecules, biology)

Sample handling (radiation damage)
Sub nanometer resolution

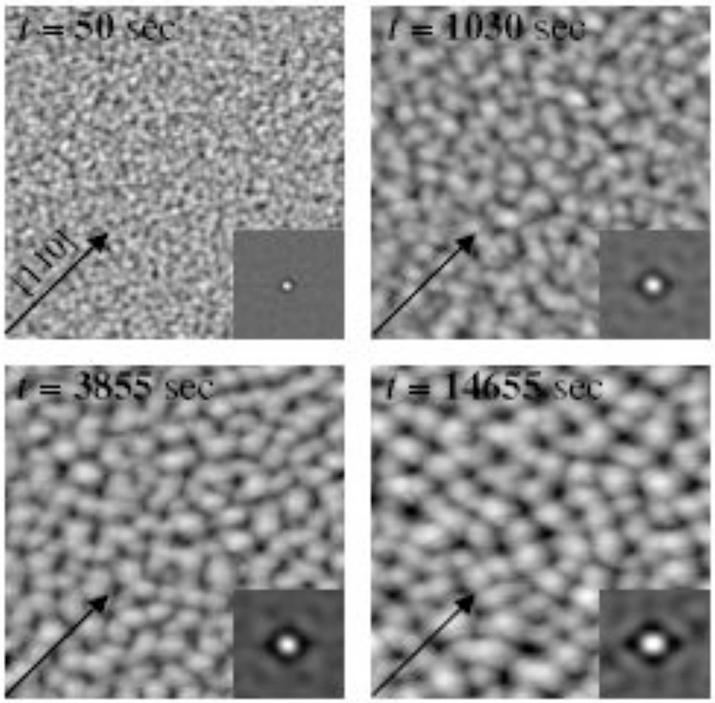


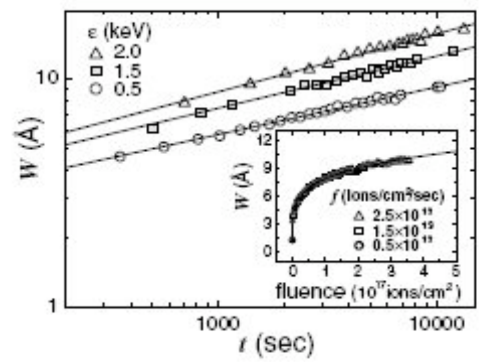
Protein crystallography
Solution small angle x-ray scattering

http://www.rekihaku.ac.jp/e-rekihaku/115/cover.html

Applications of Coherent Diffraction Imaging

Nano dot formation kinetics by ion sputtering Pd (001) surface





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Quantum dot structures

Nano-pattern

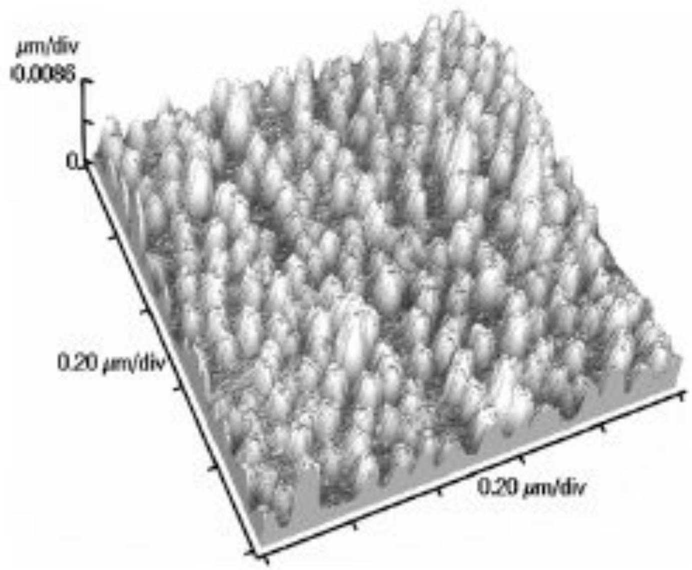

Fig. 1. AFM image of uncapped $In_{0.5}Ga_{0.5}As$ quantum dots formed on GaAs(0 0 1) surface.

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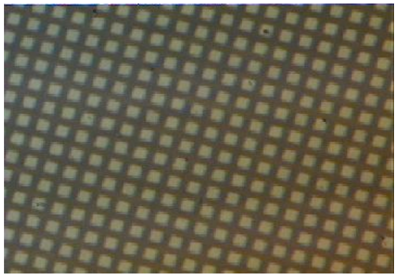

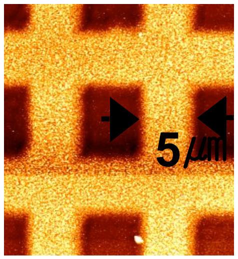

Summary

- Developed generalized oversampling algorithm
- Verified with He-Ne laser diffraction
- Achieved hard x-ray coherent diffraction imaging

  Wavelength: 0.17 nm

  Resolution ~ 20 nm Field of view ~ 2 μm
- Testing the generalized algorithm with x-ray diffraction